Effect of aeration and leachate recirculation on carbon and nitrogen removal from landfills

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ABSTRACT

Landfilling is as yet the final disposal method used in integrated waste management systems. However, it is always followed by emissions of landfill gas and leachate. The landfill gas can cause health hazards, as well as increase greenhouse effects. While landfill leachate typically contains a variety of contaminants that can leak and seriously contaminate soil, groundwater and surface water if the landfill is not well constructed and operated in accordance with the technical requirements.

In recent decades, waste management measures and landfill techniques in many places in the world, especially in developed countries have changed greatly. Sustainable landfilling has become a fundamental objective in many modern waste management concepts. “Sustainable landfill” can be understood as a landfill where the deposited waste is in a state near to stability. In this phase the remaining conversion processes are low and emission release is at the acceptable level.

A new and promising trend in solid waste management is to operate the landfills as bioreactors. Bioreactor landfills are controlled systems in which moisture addition (often leachate recirculation) and/or aeration are used to accelerate the degradation of biodegradable organic fraction of the deposited waste.

In Europe, several waste management strategies toward avoidance of waste generation, re-use of materials and resources, as well as energy utilisation of waste have been regulated to reduce the amount of landfilled waste and environmental pollution caused by landfills.

The establishment of mechanical biological treatment (MBT) concept has been followed by diverse developments of corresponding technologies. The incorporation of MBT technologies in municipal solid waste (MSW) management has greatly changed the solid waste and landfill management. MBT not only significantly reduces the quantity of the output materials to be landfilled, but also reduces a considerable proportion of organic compounds and nutrients in the output materials. It means that the output materials are much more stabilised in comparison to the input MSW. As a result, a reductions of both gas and leachate emission potentials after the emplacement of such output materials on landfills seems to be obvious.

The main difference between leachates from MBT and MSW landfills is the low levels of organic carbon and ammonium in the MBT ones. Ammonium is regarded as the most problematic parameter in leachate from MSW landfills because it is normally present at high concentrations and persistent under anaerobic condition. Ammonium level in

leachate from MBT landfills is much lower than that in leachate from MSW landfills, but is still relatively high compared to concentrations which are acceptable on the long term. Therefore, the treatment of leachate in the future is no longer or less focus on organic matter which has been dominant so far, rather on taking care of ammonium.

A part of the study was about the effect of leachate recirculation on the characteristics of leachate from MBT residue and fresh MSW. The major part of the study focused on the effect of such recirculation combining with aeration on leachate quality of such waste materials. The lab-scale study results clearly showed that recirculation of leachate alone has little effect on the characreristics of leachates from both MBT residue and fresh MSW in the experimental duration. Meanwhile, such combination technique gives a remarkable influence on improving the quality of leachates from both types of investigated materials. Very high nitrogen removal efficiency (up to 99 %) was achieved by applying this combination technique to MBT residues, whereas organic carbon components were slightly affected. At the same time, the combination technique also has strong impacts on reduction of ammonium and organic carbon substances in the leachate from MSW.

Though the combination technique results in considerable decreases of ammonium nitrogen in the leachates from both types of materials, the mechanism leading to such decreases is different. As for lab-scale lysimeters containing MBT residues, the study results indicated that nitrogen was lost via nitrification and denitrification processes by aeration. Meanwhile, the reduction of ammonium nitrogen in case of MSW was most likely due to the volatilisation of free ammonia. Despite such different mechanisms, the combination of in-situ aeration and leachate recirculation can be a simple and reliable measure to improve the quality of leachates from both MBT residue and MSW.

ZUSAMMENFASSUNG

Weltweit gesehen bildet die endgültige Ablagerung von Abfällen auf Deponien immer noch das letzte Glied im Rahmen von Abfallwirtschaftskonzepten, obwohl die Deponierung mit zahlreichen Problemen behaftet ist. Von Bedeutung sind hierbei vor allem die unkontrollierten, umweltrelevanten Gas- und Sickerwasseremissionen. Die im Deponiekörper entstehenden Gase tragen zur Klimaerwärmung bei, unkontrollierte Deponiesickerwasseremissionen können zu einer Kontamination des Bodens sowie von Grund- und Oberflächenwässern führen. Diese negativen Umweltauswirkungen können insbesondere in Schwellen- und Entwicklungsländern verstärkt auftreten, in denen die Deponiekonstruktion nicht den aufwändigen westlichen Standards entspricht.

Das Wissen um die langfristig anhaltenden potentiellen Umwelteinwirkungen hat in den letzten Jahren vor allem in den Industrieländern den Aspekt der Nachhaltigkeit bei der Abfalldeponierung in den Fokus der Betrachtungen rücken lassen. Bei einer nachhaltigen Deponierung wird davon ausgegangen, dass zwar Emissionen erfolgen, sie jedoch so gering sind, dass eine Umweltbeeinträchtigung nicht zu befürchten ist. Dieses Ziel kann durch eine Änderung des Deponiebetriebes oder durch eine Vorbehandlung der Abfälle vor der Ablagerung erfolgen.

Bei bereits abgelagerten Abfällen können durch eine gezielte und gesteuerte Bewässerung (häufig durch eine Kreislaufführung des Sickerwassers) und/oder Belüftung organische Substanzen beschleunigt biochemisch abgebaut werden, so dass das langfristige Emissionspotential reduziert wird. Die Deponie wird ähnlich einem Bioreaktor betrieben.

Weltweit wird versucht, das Abfallaufkommen und das Schadstoffpotential in den abzulagernden Abfällen durch Recycling und Behandlung der Abfälle zu reduzieren. Ein mögliches abfallwirtschaftliches Konzept hierbei ist die mechanisch-biologische Abfallbehandlung (MBA). Durch den Einsatz von den Abfällen angepassten Technologien wird die Masse der abzulagernden Abfälle sowie das in den zu deponierenden Restmassen noch vorhandene Emissionspotential stark reduziert.

Wesentliche Unterschiede in den Sickerwasserkonzentrationen von Deponien mit frischen Abfällen bzw. MBA-Material bestehen bei Kohlenstoff und Ammonium. Insbesondere bei Kohlenstoff liegen die Werte bei MBA-Deponien um Größenordnungen unter denen von Frischabfällen. Bei Ammonium sind die Werte der MBA-Deponie ebenfalls geringer, würden aber bei einem unkontrollierten Sickerwasseraustritt immer noch zu einer Umweltbeeinträchtigung führen. Ammonium ist auch in Hinblick auf die langfristige Emissionssituation bedeutend, da es unter den anaeroben

Deponiebedingungen kaum abgebaut wird. Die Ammonium-Konzentration im Sickerwasser ist daher maßgebend bei der Ermittlung von Zeiträumen, in denen eine Fassung und Behandlung der Wässer erforderlich ist.

Die vorliegende Dissertation befasst sich daher mit der Reduktion der Kohlenstoff- und Ammonium-Konzentration im Sickerwasser aus MBA-Abfällen nach der Ablagerung. Zum Vergleich werden auch frische Abfälle in die Betrachtungen miteinbezogen.

Ein Schwerpunkt der Untersuchungen liegt in der Ermittlung der Auswirkung einer Sickerwasserkreislaufführung auf die Eigenschaften der Sickerwässer. Zusätzlich zur Kreislaufführung erfolgt auch eine Belüftung des Abfalls, um Abbauprozesse zu beschleunigen. Die im Labormaßstab durchgeführten Versuche zeigen deutlich, dass eine alleinige Sickerwasserkreislaufführung bei beiden Abfällen nur einen geringen Einfluss auf die Sickerwasserqualität hat. Bei einer Kombination der Verfahren (Kreislaufführung plus Belüftung) jedoch kann innerhalb kurzer Zeiträume die Sickerwasserbelastung bei beiden Abfällen z. T. deutlich reduziert werden. Sehr hohe Wirkungsgrade von bis zu 99,9 % werden bei der Elimination von Stickstoff im MBA-Material erreicht. Bei organischem Kohlenstoff ist der Wirkungsgrad geringer, wobei hier durch die Behandlung vor der Ablagerung bereits eine Reduktion von über 90 % erzielt wird.

Obwohl die Kombinationstechnik zu einer beträchtlichen Abnahme von Ammoniumstickstoff im Sickerwasser beider Materialien führt, ist die Ursache für die Abnahme unterschiedlich. Die Lysimeter-Versuche mit MBA-Material zeigen, dass die Reduktion durch eine Nitrifikation sowie Denitrifikation als Folge der Belüftung erfolgt. Dagegen ist die Reduktion von Ammoniumstickstoff bei frischen Abfällen höchstwahrscheinlich durch der Verflüchtigung von freiem Ammoniak zu Stande gekommen. Trotz dieser unterschiedlichen Mechanismen, kann die Kombination von in-situ-Belüftung und einer Kreislaufführung des Sickerwassers eine einfache und zuverlässige Methode darstellen, um die Qualität des Sickerwassers aus MBA-Rückstand und Siedlungsabfall langfristig zu verbessern.

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ABBREVIATIONS AND DEFINITIONS

Abbreviations

BOD_5	Biochemical Oxygen Demand after 5 days
COD	Chemical Oxygen Demand
DIN	Deutsche Industrie Norm (German Industrial Standard)
DM	Dry Matter
DNRA	Dissimilatory Nitrate Reduction to Ammonium
DOC	Dissolved Organic Carbon
EC	Electrical Conductivity
EEA	European Environment Agency
EU	European Union
ISO	International Organisation for Standardisation
LSR	Landfill Simulation Reactor(s)
MBT	Mechanical Biological Treatment
MSW	Municipal Solid Waste
NH_4-N	Ammonium nitrogen (NH_4-N ≅ NH_4^+/1.3)
NMOCs	Nonmethane organic compounds
NO_3-N	Nitrate nitrogen (NO_3-N ≅ NO_3^-/4.4)
NO_2-N	Nitrite nitrogen (NO_2-N ≅ NO_2^-/3.3)
RI_4	Respiratory activity Index after 4 days
TKN	Total Kjeldahl Nitrogen (= NH_4-N + org-N)
TN	Total Nitrogen (= TKN + NO_3-N + NO_2-N)
TOC	Total Organic Carbon
VFA	Volatile Fatty Acids

Definitions

- Acetogenesis: A biological reaction wherein volatile fatty acids are converted into acetic acid, carbon dioxide, and hydrogen.

- Acidogenesis: A biological reaction wherein simple monomers are converted into volatile fatty acids.

- Aerobic microbes/ conditions: Microbes that require the presence of air or free oxygen for life/ conditions in which air or free oxygen exist.

- Anaerobic bacteria/ conditions: Bacteria that require the absence of free oxygen for life/ conditions in which air or free oxygen is absent.

- Anoxic: Severe deficiency of oxygen.

- Biochemical Oxygen Demand (BOD): The amount of dissolved oxygen needed by aerobic biological organisms in a body of water (wastewater) to break down organic material present in a given water sample at certain temperature over a specific time period. BOD is used as an indirect measure of biologically degradable material present in wastewater. This is not a precise quantitative test, although it is widely used as an indication of the organic quality of water. The BOD_5 value is most commonly expressed in milligrams of oxygen consumed per litre of sample during 5 days of incubation at 20 °C and is often used as a robust surrogate of the degree of organic pollution of water.

- Chemical Oxygen Demand (COD): A measure of the oxygen required to oxidise all compounds, both organic and inorganic, in wastewater. COD is typically measured in mg/L, which indicates oxygen consumed per liter of solution.

- Electrical conductivity (EC): The reciprocal of electrical resistivity, and measures the ability of a material to conduct an electric current.

- Flushing: In the context of this dissertation, flushing is referred to the process in which a comparatively large volume of the liquid phase (water at the beginning and then leachate) passes through the solid phase (waste). This process should occur in a relatively short period of time.

- Hydrolysis: A chemical reaction where particulates are solubilised and large polymers converted into simpler monomers;

- Landfill: A site for the disposal of waste materials by burial. Historically, landfills have been the most common methods of organised waste disposal and remain so in many places around the world. Today, landfill sites are constructed and operated to strict technical standards in order to reduce environmental effects.

- Landfill Simulation Reactor (LSR): A reactor which is used to simulate certain conditions and/or processes of a landfill.

- Leachate: The whole liquid, including rain precipitation, water content of the waste itself and water produced by biological processes that percolates through the landfill body.

- Leaching: In the context of this dissertation, leaching is referred to the process in which either a small volume of liquid phase (external sources) percolates through the solid phase in a relatively long period of time or the liquid from the solid waste itself (internal sources) seeps down to the bottom of the container. It can be understood that as a certain volume of liquid phase is pumped into the surface of a solid phase, flushing occurs prior to leaching.

- Mechanical Biological Treatment (MBT): A technique which combines mechanical sorting and separation with a form of biological treatment such as composting or anaerobic digestion.

- Methanogenesis: A biological reaction where acetates are converted into methane and carbon dioxide, while hydrogen is consumed.

- Respiratory Index after 4 days (RI_4): The measure for the aerobic biodegradation of the organic content of the waste. Here, the oxygen consumption is measured in 4 days.

- Total nitrogen (TN): The sum of all forms of nitrogen in a liquid solution, including inorganic nitrogen (nitrate, nitrite, ammonia-N) and organic forms of nitrogen.

- Total Organic Carbon (TOC): A sum measure of the concentration of all organic carbon atoms covalently bonded in the organic molecules of a given liquid or solid sample.

- Washing: In the context of this dissertation, washing has the same meaning as flushing, but requires much more water and/or liquid volume than in the case of flushing.

CHAPTER 1: INTRODUCTION

1.1 Background and motivation

Between 2011 and 2050, the world population is expected to increase by 2.3 billion, passing from about 7.0 to 9.3 billion (United Nations, 2011). At the same time, the population living in urban areas is projected to gain 2.6 billion, passing from 3.6 billion in 2011 to 6.3 billion in 2050. In particular, Asia is projected to see the highest urban population increase by 1.4 billion, followed by Africa with an increase by 0.9 billion (United Nations, 2012). Rapid increase in volume and types of solid and hazardous waste as a result of continuous economic development, industrialisation, as well as urbanisation taking place around the world has become a serious issue. Therefore, the importance of having an efficient and effective solid waste management system is more important than ever before.

Landfilling is so far still the most common waste management solution all over the world. This technique seems to offer a fast and simple way for waste management at reasonable costs. However, the landfilling management measures in many developing countries are very poor (e.g. most of MSW is yet either disposed of by open dumping, open burning, controlled burning and tipping, or directly landfilled without any pre-treatment). Moreover, landfilling is the opposite of sustainability in several respects. It not only misspends the valuable resources in the deposited refuse, but also brings about potential long-term health hazards and environmental burdens (Ettler et al., 2008; Öman and Junestedt, 2008). Landfilling of untreated MSW can bring on several environmental problems that may last for hundreds of years. Biodegradation of organic matters in landfills produces landfill gases which partially contribute to the greenhouse effects. Bad odours from landfill and landfill leachate can be spread out to the surroundings. Landfill leachate can also carry several pollutants to groundwater systems, especially in case that the bottom liner system is not present or not well constructed. Environmental problems related to leachate can include groundwater pollution and oxygen depletion and ecotoxicity in surface waters (Kjeldsen et al., 2002; Pablos et al., 2011). In many European countries, not to mention the rest of the world, this is still the case (EEA, 2009; Laner et al., 2012). However, those problems are even more common and severe in developing countries. The current situation calls for more advanced techniques to avoid negative effects of landfilling.

Many management strategies toward avoidance of waste generation, re-use of materials and resources, as well as energy utilisation of waste have been regulated to reduce the amount of landfilled waste and environmental pollution caused by landfills. During

recent decades, the European Union (EU) has issued several ordinances on waste management and landfilling (EEA, 2009). Of which, the most important legislation for landfills is the Waste Framework Directive (75/442/EEC amended, later replaced by 2008/98/EC). The Directive with its hierarchy of waste management options stated that prevention and minimisation of wastes are of highest priority, followed by recovery, reuse and recycling. The EU Landfill Directive (1999/31/EC) requires, among other things, that all waste must be pre-treated prior to landfilling, and restricts the landfilling of organic waste. The Landfill Directive has been effective, advancing the diversion of waste from landfills and increasing the use of alternative waste management options. The diversion of waste from landfills has led to the closure of many landfill sites in Germany and elsewhere (EEA, 2009). The types of waste going to landfills have therefore also changed. The concentration of organic matter has decreased and its degradability is reduced thanks to pre-treatment (EEA, 2009). For example, it was stated in the Directive that share of biodegradable landfilled MSW has to be reduced by 25 % before the year 2006, compared to the amount of biodegradable waste in 1994 and, further, by at least 50 % before 2009 and 65 % before 2016. The Directive also sets requirements and criteria on the construction of landfills, in which all landfills must have a natural or constructed geological barrier to prevent exchange of water with the surroundings. Together with an increasing interest in the utilisation of resources found in waste, such as energy and recyclable materials, the new regulation has led to a renovation of waste management in general and landfill management in particular.

Germany was the first country in the EU to introduce producer responsibility with a packaging waste regulation in 1991, in which the producer of a product is generally responsible for the product when it becomes waste. Germany was also among the first European countries to introduce policies, including schemes for collecting packaging waste, biowaste and waste paper separately, to limit landfilling in the 1990s. The result of this was that Germany already recycled about 48 % of MSW by 2001, meanwhile approximately 25 % was landfilled and 22 % was incinerated. In 2010, the level of recycling increased to 62 %, landfilling of untreated MSW was near to 0 % and incineration increased to 37 % (EEA, 2013). However, this increase of incineration was not a real reflection of the actual amount of MSW incinerated. According to the German reporting to Eurostat, “incineration of MSW (without energy recovery)” includes "treatment for disposal", mostly referring to MBT” (Eurostat, 2012). Moreover, the waste generated during the pre-treatment process such as sorting or MBT will also include waste ending up partly in incinerators and in landfills and the latter part of this waste is not necessarily reported as landfilled but as incinerated (EEA, 2013). In a study of the flows from MBT in Germany by Thiel and Thomé-Kozmiensky (2011), it was stated that

22 % of the input of approximately 6.4 million tonnes into MBT plants ended up in landfills in 2007.

The changes in waste management have also changed potential environmental impacts (Manfredi et al., 2010). In landfills pursuant to the new legislation, gas formation may be negligible, then posing leachate as the main emission pathway. The decrease in organic content, therefore, also means that inorganic pollutants, such as heavy metals, ammonium may become relatively more pronounced in the leachate.

It is well known that landfill leachate is a very complex mixture of many different types of pollutants with varying concentrations. The main environmental problems at landfill sites are the infiltration of leachate and its subsequent contamination of the surrounding land and aquifers if the landfills have no bottom liner system and/or the bottom liner does not meet technical standards.

In Germany and many other developed countries, the utilisation of MBT technologies for MSW management have resulted in marked improvements regarding organic matter and nutrients of the output materials going to landfills and their leachates produced. However, ammonium concentration of the leachate is still relatively high comparing with the discharge limits.

1.2 Objectives and scope of the research

The objective of this research is to investigate whether either leachate recirculation regime or a combination of this regime with aeration can improve the quality of leachate released from MBT residue and MSW. The research is based on experimental works, in which a series of laboratory lysimeters were used. Such lysimeters were loaded with either MBT residue or MSW and operated under different boundary conditions to simulate different types of bioreactor landfills. The leachates generated from those lysimeters were characterised by varying parameters, in which nitrogen and organic carbon were particularly investigated and examined above others. The focus was set on organic carbon and nitrogen parameters because of the following reasons:

- Changes in the quantity and quality of such parameters are among the major changes expected in landfills due to the differences in waste management;
- Organic carbon compounds in the deposited waste and its leachate strongly influence the stabilisation process of a landfill;
- Ammonium is likely to be among the pollutant groups of most long-term concern in landfill leachates.

The research focused on leachate emissions from MBT residues and MSW collected from an MBT plant in Germany, in which anaerobic fermentation is followed by aerobic stabilisation. MSW is the input material, whereas MBT residue is the output of MBT process. Such waste materials are within the legal framework of Germany, but are also relevant for wastes in other parts of the world to a large extent. For review purpose, German landfills and their data are of focus, but landfills from other countries were included for comparison purposes.

1.3 Structure of the dissertation

This dissertation is constructed into seven chapters. After the introduction in chapter 1, the literature review addressing the principle relating to degradation of waste, nitrogen conversion processes, and emission behaviours of landfills is presented in chapter 2. Chapter 2 ends up with the statement of problem. Thereafter, a description of the materials, laboratory settings, analytical methods and boundary conditions used as well as the calculation is presented in chapter 3. The experimental works undergo three phases (1, 2, and 3), in which the laboratory settings and boundary conditions are specific for each phase. The next main chapters 4, 5, and 6 are constructed basing on the experimental results achieved from phase 1, 2, and 3, respectively. In each main chapter, the experimental results are summarised, evaluated, and discussed as well as compared with the previous study data. Finally, conclusions, recommendations, and outlooks are presented in Chapter 7.

CHAPTER 2: LITERATURE REVIEW

2.1 Principles of biochemical and microbial conversion processes in landfills

2.1.1 Aerobic degradation

The aerobic degradation of organic matters by microorganisms is mainly implemented in the presence of oxygen to form carbon dioxide, water, salts, humic substances and organisms mass (Schlegel, 1992; Krogmann, 1994).

The first step of biological degradation is hydrolysis, in which the natural macromolecular substances are broken down by exoenzymes into their monomeric constituents. The monomers sugars, amino acids and fatty acids can then be taken up by the microorganisms, incorporated into the cell substance or mineralised under oxygen consumption to gain energy. By the mineralisation of amino acids, the monomeric constituents of proteins are also generated in addition to ammonia gas. The end products of a complete mineralisation are carbon dioxide and water.

The aerobic degradation of organic substances generally takes place in short duration due to the high oxygen demand of waste relative to the limited quantity of oxygen present inside a landfill. In this stage, there is usually no substantial leachate generation.

In very old landfills, where organic matters in the deposited wastes mostly consist of the more refractory organic carbon substances, a second aerobic phase may appear in the upper layer of the landfill. In this phase the methane production rate is very low and air starts diffusing from the atmosphere, giving rise to aerobic zones and zones with redox potentials too high for methane formation (Christensen and Kjeldsen, 1989).

During aerobic degradation, it is important to keep the water content at a suitable level in addition to the sufficient supply of oxygen to the microorganisms for an optimal degradation. According to Heerenklage et al. (1994), the maximum water content of refuse should not exceed 55 to 65 % on a wet basis. The research findings of Rees (1980b) suggest that the landfill gas production rate rises exponentially with increase in water content up to 60% on a wet basis. Water contents higher than 60% do not seem to enhance nor decrease the gas production rate (Pohland and Harper, 1986). However, low water content inhibits the growth of microorganisms. Microbial activity no longer takes place at water contents below about 12 - 25 % on a wet basis (Bidlingmaier, 1983; Schuchardt, 1988; and Golueke, 1989). Hartz and Ham (1983) reported that methane production would decrease as water content in the waste reduces and would cease completely if the water content is below 10 % on a wet basis.

2.1.2 Anaerobic degradation

Anaerobic degradation involves a series of processes in which microorganisms break down biodegradable materials in the absence of oxygen. Acetogenic bacteria then convert these resulting organic acids into acetic acid, along with additional ammonia, hydrogen, and carbon dioxide. Finally, methanogenic bacteria convert these products to methane and carbon dioxide.

The complete anaerobic degradation of organic matter takes place in four phases, in which three different types of microorganisms are involved (Figure 2.1).

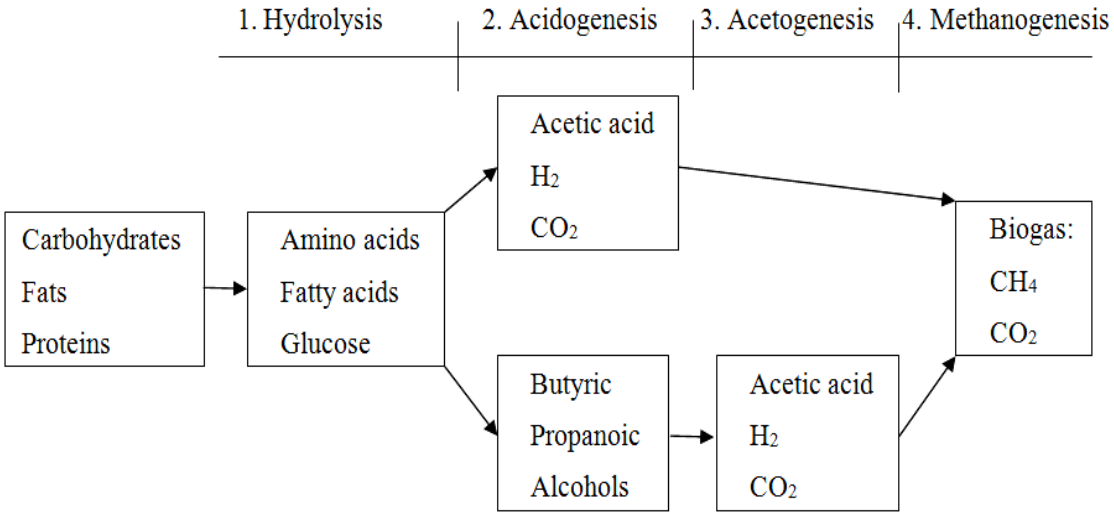


Figure 2.1. Reaction steps in the anaerobic fermentation process (adapted from Faulstich et al., 1995)

The degradation process begins with bacterial hydrolysis of the input materials to break down insoluble organic polymers, including carbohydrates, fats, proteins and so that they can be used by other bacteria and/or microorganisms. In the subsequent phase, the acid fermentation - or acidogenesis, acidogenic bacteria (fermentative bacteria) convert the sugars and amino acids into organic acids, alcohols, hydrogen and carbon dioxide. While acetic acid, hydrogen and carbon dioxide can directly enter into the conversion processes of the last phase - methanogenesis, other remaining organic acids, alcohols and carbon dioxide will be converted into acetic acid and hydrogen during acetogenesis phase by acetogenic bacteria.

In methanogenesis phase, acetic acid, hydrogen and carbon dioxide are converted to methane by methanogenic bacteria. The total carbon dioxide amount cannot be fully converted because of limited amounts of hydrogen, so that carbon dioxide is obtained in addition to methane at the end of the complete anaerobic degradation. Besides these two

main gas components, small quantities of other gases such as ammonia and hydrogen sulphide can also be enclosed in the biogas. They arise essentially due to anaerobic degradation of protein (Zachäus, 1995).The complete anaerobic mineralisation is a highly complex process, since the fermentative bacteria have propagation rates much greater than the acetogenic and methanogenic bacteria.

The anaerobic conversion process is significantly influenced by the substrate temperature and pH value.

Research suggests that anaerobic processes are optimised when the waste is within either mesophilic (30 to 38 °C) or thermophilic (50 to 60 °C) temperature range (Parkin and Owen, 1986). According to Mudrack and Kunst (1991), the optimum temperature for the acid formation is about 30 °C, meanwhile the methanogenic bacteria predominantly exist at the range from 30 ° to 40 °C. Nevertheless, there are also thermophilic methane bacteria that their optimum temperature ranges from 55 to 65 °C. The higher thermophilic temperatures enhance the rate at which organic matter is converted to volatile organic acids but lead to a lower yield of methane compared to the lower mesophilic temperatures (Pohland et al., 1993). Methanogenic bacteria are very sensitive to temperature changes. This is particularly the case for thermophilic microorganisms. However, temperature variations of ± 3 °C in the mesophilic range have no major influence on the anaerobic degradation (Winter, 1985).

The optimal pH values for anaerobic systems range between 6.8 and 7.4 (Parkin and Owen, 1986). Initially, the leachate pH values may be neutral, but they are generally dropped after the onset of anaerobic conditions, particularly during the acid forming phase due to the accumulation of volatile fatty acids in the leachate. However, the pH values will eventually increase to neutral conditions as methanogens consume these acids (U.S EPA, 2006). Due to the different pH optimal values in the acid-forming (pH from 5.3 to 6.7) and the methanogenic microorganisms (pH from 6.8 to 7.2), the whole process is only obtained for a narrow pH range, in which all microorganisms involved in converting satisfactory substrate. A strong inhibition of methane formation begins at pH values below 6 (Präve et al., 1984).

2.2 Nitrogen transformation processes and the fate of nitrogen in landfills

Nitrogen is one of the most abundant elements in the tissues of all organisms and is a component of many biochemicals, particularly amino acids, proteins, and nucleic acids. Consequently, nitrogen is one of the critically important nutrients and is required in relatively large quantities by all organisms (Bisen et al., 2012).

Nitrogen exists in many organic and inorganic forms in the environment. Organic nitrogen encompasses a diversity of nitrogen-containing organic molecules, ranging from simple ones such as urea, amino sugars, amino acids, nucleic acids, and proteins to large and complex molecules such as the humic substances. Inorganic nitrogen includes a variety of substances, in which nitrogen can exist in liquid forms (ammonium, nitrate, nitrite) or gaseous ones (ammonia, dinitrogen, nitric oxide, and nitrous oxide).

Nitrogen can exist in several oxidation states, ranging from -3 in the most reduced forms to +5 in the most oxidised forms (Figure 2.2).

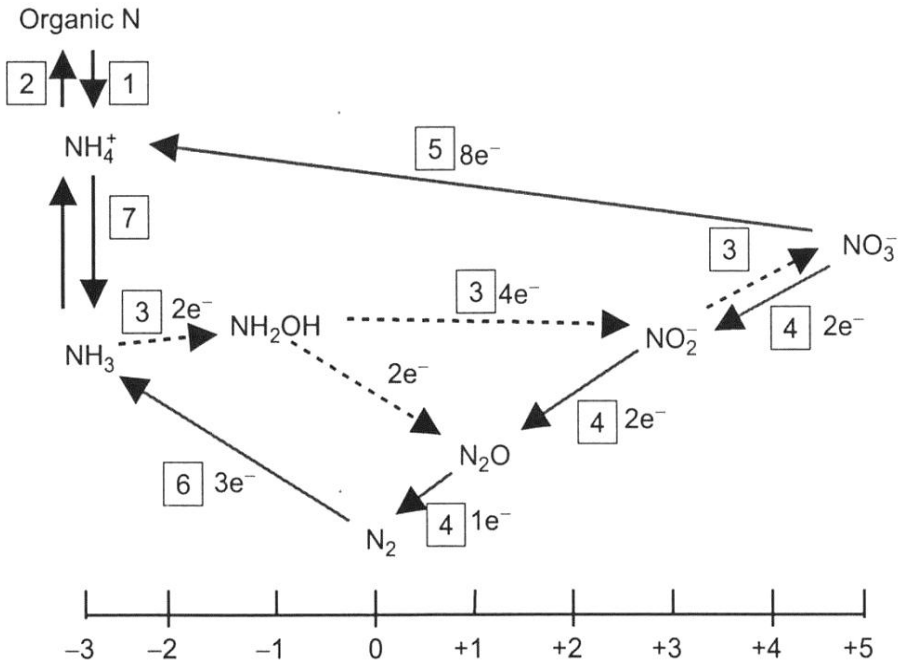


Figure 2.2. Oxidation state and transformation pathways of nitrogen (Reddy and DeLaune 2008)

Pathway 1 indicates ammonification, which is the first step in mineralisation of organic nitrogen. Pathway 2 shows immobilisation, whereby ammonium is assimilated into the biomass of plants and/or microbes. Pathways 3 and 4 show nitrification and denitrification, respectively. Pathway 5 indicates nitrate reduction to ammonia nitrogen. The reduction of nitrate to ammonia gas is referred to as dissimilatory nitrate reduction, whereas the nitrate reduction to ammonium is defined as assimilatory one. Pathway 6 shows nitrogen fixation, which is the reduction of atmospheric, inert nitrogen to ammonia nitrogen that plants and microbes can then use. Pathway 7 shows ammonia volatilisation, a physico-chemical process controlled by the pH of the environment.

2.2.1 Ammonification, volatilisation, and ammonium sorption processes

The organic nitrogen within MSW mainly consists of proteins (Burton and Watson-Craik, 1998; Vigneron et al., 2007). The ammoniacal nitrogen in leachate is generated from the organic nitrogen content of the deposited waste. The conversion of organic nitrogen to ammoniacal nitrogen by microorganisms is termed ammonification. Ammoniacal nitrogen produced by ammonification is dissolved in the leachate and may be transformed and/or removed via different pathways, such as volatilisation, sorption, or biological processes when in an aerobic environment (Berge et al., 2005).

Volatilisation only occurs when free ammonia is present. In aqueous phase, there is a chemical equilibrium between ammonium ion and free ammonia via ammonium hydroxide (Eq. 2-1).

$$NH_4^+ + OH^- \leftrightarrow NH_4OH \leftrightarrow NH_3 + H_2O \qquad \text{(Eq. 2-1)}$$

The position of the ammonium-ammonia equilibrium depends on the pH and temperature of the aqueous phase (Eq. 2-2, according to Anthonisen et al., 1976).

$$[NH_3\text{-}N] = [NH_4^+\text{-}N] * \frac{10^{pH}}{(e^{(6344/(273+T))} + 10^{pH})} \qquad \text{(Eq. 2-2)}$$

At temperatures of 55 °C and a pH value of 8.5, ammonium and ammonia are present in approximately equal proportions. At pH levels above 10.5 to 11.5, the majority of the ammoniacal nitrogen present in solution is in the form of free ammonia.

Several compost studies showed that losses of nitrogen by NH_3 volatilisation were significant at pH values above 7.0 and temperatures above 40 °C (Bishop and Godfrey, 1983; Witter and Lopez-Real, 1988; Tiquia and Tam, 2000; Sanchez-Monedero et al., 2001).

Air flow plays an important role in volatilisation process occurring in landfills. As air is introduced, removal pathways for volatilisation of dissolved free ammonia are created so that free ammonia is able to leave the landfills. Air flow also dilutes the concentration of free ammonia above the leachate, therefore increasing the driving force for dissolved free ammonia to partition to the gaseous phase (Thomas, 1982; Henry et al., 1999).

A study on emission behaviours of aerated landfills using a lab-scale simulated aerobic bioreactor landfill by Ritzkowski and Stegmann (2003) stated that 50 % of the ammoniacal nitrogen initially presents in the leachate is volatilised at a pH of 7.4 and a temperature of 35 °C.

Sorption of ammoniacal nitrogen to waste may be significant in bioreactor landfills because ammonium is commonly present at high concentrations. According to Laima (1994), ammonium is known to sorb onto various inorganic and organic compounds. Moreover, Nielson (1996) stated that the amount of ammonium sorbed on some organics exceeds the mass found in the bulk liquid. Sorption of ammonium to the waste allows the temporary storage of ammonium before it being used in other processes, such as nitrification and volatilisation, and may also result in the slow dissolution of ammonium over time (Heavey, 2003). Ammonium sorption depends on several factors, including pH, temperature, ammonium concentration, and ionic strength of the bulk liquid. According to Nielson (1996) and Heavey (2003), sorption of ammonium tends to decrease as ionic strength of the bulk liquid increases due to ion-exchange effects. The sorbed ammonium is released and exchanged with other ions in the bulk liquid, particularly with those having higher concentration or selectivity. The addition of sodium or potassium sulphate solution can be used to desorb ammonium from the solid waste (Berge et al., 2005).

2.2.2 Nitrification

Nitrification is generally known as the autotrophic conversion (biological oxidation) of ammonium to nitrite, and finally nitrate by nitrosomonas and nitrobacter bacteria groups (Eqs. 2-3 and 2-4).

$$NH_4^+ + 1.5\ O_2 \rightarrow NO_2^- + 2\ H^+ + H_2O \qquad \text{(Eq. 2-3)}$$

$$NO_2^- + 0.5\ O_2 \rightarrow NO_3^- \qquad \text{(Eq. 2-4)}$$

In a broader sense, nitrification is defined as the conversion of organic or inorganic compounds from reduced state to a more oxidised state (Körner, 2008). It also covers the activities of heterotrophic nitrifiers. However, the up-to-date knowledge of heterotrophic nitrification is limited in comparison to that of autotrophic nitrification.

The nitrifying chemotrophic autotrophs involved (nitrifiers) must fix and reduce inorganic carbon (mostly from carbon dioxide) to use as their carbon source for cell synthesis (Rittman and McCarty, 2001). They obtain energy for reduction of carbon dioxide through the oxidation of nitrogen compounds. These organisms cannot use organic carbon as the sole carbon source for growth or obtain energy by oxidising substrates other than those containing nitrogen. Meanwhile, heterotrophic nitrifiers use organic substances as the energy source and gain no energy from the oxidation of ammonium (Reddy and DeLaune, 2008).

Possible intermediate compounds can be formed during the oxidation of ammonium to nitrate (Figure 2.3).

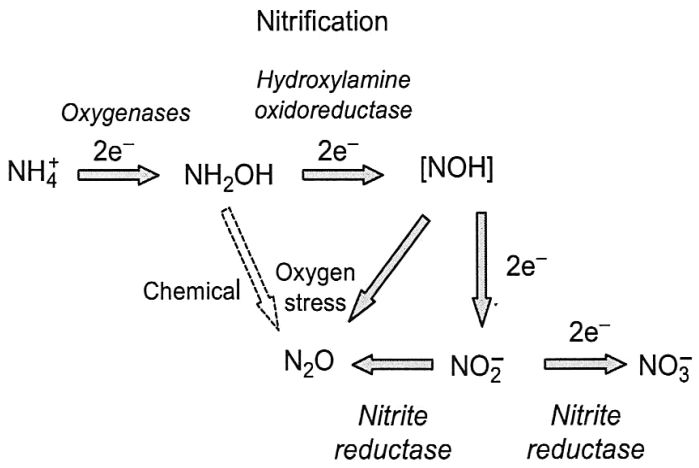


Figure 2.3. Pathways and intermediate products during nitrification (Reddy and DeLaune, 2008)

Nitrification is almost nonexistent in conventional landfills, as well as in bioreactor landfills in which air is not added. However, in landfills in which air is purposely added, nitrification can be a significant pathway for nitrogen removal (Berge et al., 2005).

2.2.3 Denitrification

According to Rittman and McCarty (2001), denitrification is an anoxic process that reduces nitrate to nitrite, nitric oxide, nitrous oxide, and finally to nitrogen gas (Eqs. 2-5, 2-6, 2-7, and 2-8).

$$NO_3^- + 2\ e^- + 2\ H^+ \rightarrow NO_2^- + H_2O \qquad \text{(Eq. 2-5)}$$

$$NO_2^- + e^- + 2\ H^+ \rightarrow NO + H_2O \qquad \text{(Eq. 2-6)}$$

$$2\ NO + 2\ e^- + 2\ H^+ \rightarrow N_2O + H_2O \qquad \text{(Eq. 2-7)}$$

$$N_2O + 2\ e^- + 2\ H^+ \rightarrow N_2 + H_2O \qquad \text{(Eq. 2-8)}$$

Typically, denitrifying bacteria are heterotrophic, facultative aerobes, which use nitrate as an electron acceptor when oxygen is limiting or absent (Berge et al., 2005). However, many denitrifying bacteria can use nitrite, nitric oxide, or nitrous oxide, instead of nitrate, as terminal electron acceptors. Alternatively, these intermediates may release during denitrification of nitrate under unfavourable conditions, as were observed in soil (Conrad, 1996).

According to Schön et al (1994), nitric and nitrous oxides can be formed if surplus nitrate is supplied and hydrogen donors are insufficiently available. Another condition for nitrous oxide formation is when pH values are below 7.3, at which nitrogen oxido-reductase is inhibited (Knowles, 1982).

Nuske (1983) stated that biodegradable carbon must be available in the denitrification process, and denitrification decreases with increasing decomposition or humification degree. The simultaneous consumption of carbon and nitrate without requiring oxygen input is a potential advantage of denitrification (Grady et al., 1999).

$$5\ C + 4\ NO_3^- + 4\ H^+ \rightarrow 2\ N_2 + 2\ H_2O + 5\ CO_2 + Energy \qquad \text{(Eq. 2-9)}$$

Several intermediate compounds can be formed during nitrate reduction to nitrogen gas (Figure 2.4).

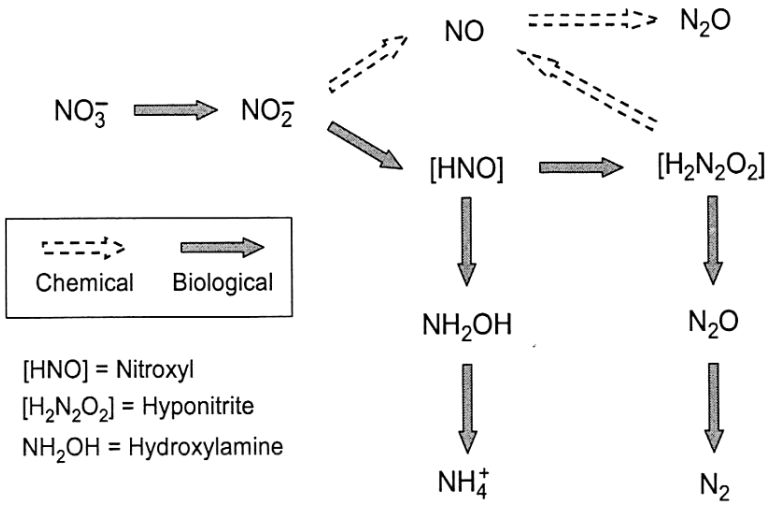


Figure 2.4. Pathways and intermediate products during nitrate reduction reaction (Reddy and DeLaune, 2008)

Nitroxyl (HNO) and hyponitrous acid ($H_2N_2O_2$) are some of the possible intermediates during denitrification. These compounds are unstable and are rapidly reduced to nitrous oxide (Reddy and DeLaune, 2008). The gaseous intermediates nitric and nitrous oxides can escape from the system and emit into the environment. Since one mole of hydrogen ion is consumed per mole of nitrate, an increase in pH is to be expected during denitrification.

Thermodynamically, in the absence of oxygen, nitrogen oxides are the most preferred electron acceptors by facultative bacterial groups. A wide range of bacteria are capable of using nitrogen oxides as electron acceptors. In which, some organisms are capable of

following through the entire reduction pathway, whereas others are capable of catalysing only one or two steps of the pathway.

Partial denitrification by certain groups of bacteria may be due to unavailability of nitrate, inability to synthesise nitrogen oxide reductases, or environmental factors such as pH, oxygen concentration, or concentration of intermediate compounds.

Nitrifier denitrification refers to reduction of nitrite to nitrous oxide in aerobic cultures formed during nitrification (Figure 2.5) by ammonium-oxidising bacteria, including chemoautotrophs, methanogens, and some heterotrophs. All of these groups are capable of reducing some of the intermediate compounds such as hydroxylamine (NH_2OH) or nitrite to nitrous oxide, especially under low oxygen conditions (Wrage et al., 2001; Zehr and Ward, 2002; Sutka et al., 2006).

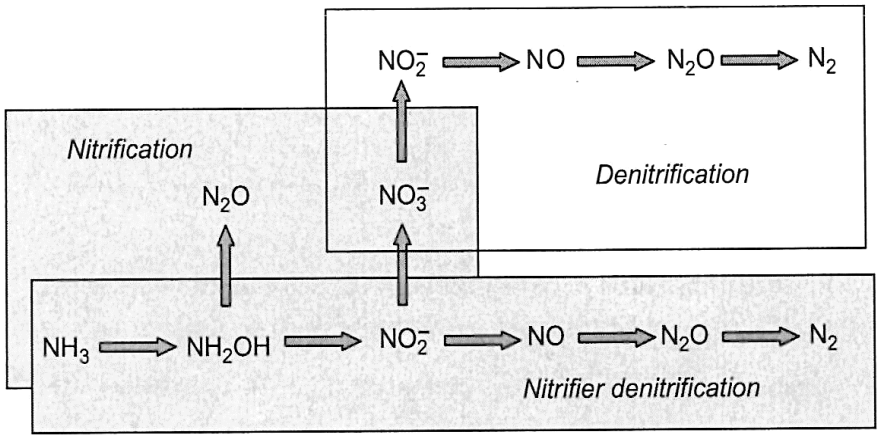


Figure 2.5. Schematic of pathways showing denitrification during nitrification (Wrage et al., 2001)

Aerobic denitrification (or co-respiration) is the simultaneous use of both oxygen and nitrogen as oxidising agents, performed by various genera of microorganisms (Robertson and Kuenen, 1984). It was classically thought that denitrification would not occur in the presence of oxygen since there seems to be no energetic advantage to using nitrate as an oxidant when oxygen is available. This process differs from anaerobic denitrification not only in its insensitivity to the presence of oxygen, but also in its higher potential to create the harmful byproduct nitrous oxide (Lloyd, 1993).

Chemodenitrification refers to abiotic conversion of ammonium to nitrite or reaction of nitrite itself with organics such as amines and inorganics such as metals, resulting in conversion of nitrite to gaseous end products. This alternative pathway has been documented in agricultural soils and marine sediments (Nelson, 1982; Van Cleemput and Baert, 1984).

Another type of denitrification is called autotrophic denitrification, which involves the Thiobacillus denitrificans. They use an inorganic sulphur source (i.e., hydrogen sulphide, sulphur, and sulphite) rather than an organic carbon source when reducing nitrate to nitrogen gas (Onay and Pohland, 2001) according to the following reaction:

$$8\ NO_3^- + 5\ HS^- + 3\ H^+ \rightarrow 4\ N_2 + 5\ SO_4^{2-} + 4\ H_2O \qquad \text{(Eq. 2-10)}$$

2.2.4 Anammox

Biological oxidation of ammonium under anaerobic conditions by nitrite is termed the Anammox process (anaerobic ammonium oxidation). Bacteria capable of Anammox use ammonium as the electron donor and nitrite as the electron acceptor (Eq. 2-11).

$$NH_4^+ + NO_2^- \rightarrow N_2 + 2\ H_2O + \text{Energy} \qquad \text{(Eq. 2-11)}$$

The Anammox process is generally favourable in environments in which retention time is long, operation is stable, nitrite is present, and electron donors that would cause nitrite reduction via denitrification are absent. It could incidentally take place simultaneously with nitrification process because of the possibility for anaerobic zones within aerobic landfills. Therefore, this process could be potential for the removal of ammonium in addition to nitrification. However, the growth rates of the Anammox bacteria are extremely slow, which leads to slow removal of ammonium as well (Berge et al., 2005).

2.2.5 Assimilatory and dissimilatory nitrite or nitrate reduction to ammonium

When nitrite or nitrate ions are reduced to ammonium ions inside the bacterial cell, the nitrogen in the ammonium ions is incorporated into cellular material. This reduction of nitrogen is termed “assimilatory” nitrite or nitrate reduction.

Dissimilatory nitrate reduction to ammonium (DNRA) refers to an anaerobic microbial pathway of the nitrogen cycle that transforms nitrate first to nitrite, and then to ammonium. DNRA in anaerobic or anoxic environments may take place in landfills according to Eqs. 2-12 and 2-13.

$$NO_3^- + 2\ H^+ + 4\ H_2 \rightarrow NH_4^+ + 3\ H_2O \qquad \text{(Eq. 2-12)}$$

$$2\ CH_2O + NO_3^- + 2\ H^+ \rightarrow 2\ CO_2 + H_2O + NH_4^+ \qquad \text{(Eq. 2-13)}$$

DNRA is favoured over denitrification in anaerobic and anoxic environments with a high chemical oxygen demand to nitrate (COD/NO_3^-) ratio. The reason is that in an electron acceptor limiting environment it is more advantageous for the microorganisms to

metabolise nitrate to ammonium and gain 8 electrons per mole of nitrate than denitrify and gain only 5 electrons per mole of nitrate (Tiedje, 1988).

The microbes responsible for the DNRA process are different from denitrifiers in that they are generally fermentative, using nitrate as electron sink, rather than respiratory and using nitrate as a terminal electron acceptor (Tiedje, 1988; Cole, 1990).

2.2.6 Nitrogen immobilisation

Nitrogen immobilisation refers to the short-, medium-, or long-term integration of inorganic nitrogen to organic nitrogen-containing compounds. It is a contrary process of nitrogen mineralisation (ammonification). Nitrogen immobilisation is an assimilative process related to microbial anabolic activity. A high amount of organic matter in the substrate should result in immobilisation. Microbial immobilisation of ammonium nitrogen depends on the carbon-to-nitrogen (C/N) ratio of organic residues undergoing decomposition (Reddy and DeLaune, 2008).

2.3 Emission behaviour of landfills

Emission behaviour of a landfill may significantly vary depending on the characteristics of landfilled waste, surrounding hydro-geological and climatic conditions, landfill age, operation regime, etc. However, the content of organic matters in the landfill plays the key role in governing the emission behaviour.

2.3.1 Organic matters in landfills

In landfilled waste, organic substances are found in many waste fractions such as organic waste, wood, paper, plastics, etc.

Under landfill conditions, such substances are either biologically inert or microbially degraded, leading to the formation landfill gas and several compounds found in landfill leachate. The more biodegradable carbon is included in landfilled waste, the more concentrated leachate and landfill gas may arise. These emissions can definitely contribute to pollute groundwater and to increase greenhouse effect. Therefore, one of the essential targets in waste management measures is to mitigate as much as possible biodegradable carbon in landfills.

In many developed countries, the application of source separate collection has been established, in which bio-waste is separated from others at the source accordingly. This measure was first applied in Witzenhausen, Germany in 1982 and was started in Austria

at the end of 80s. The subsequent development of MBT techniques in the following years have significantly contributed to the reductions of organic matters being landfilled.

Despite the source separate collection and the deployment of MBT, concentration of organic matters in the MBT residue and its leachate is still high in comparison with those regulated in the German ordinance on requirements for the discharge of wastewater into waters.

The emission behaviours are discussed for MSW and MBT landfills based on literature data. However, data for MBT landfills are limited and/or insufficient due to the fact that up to now there are actually nearly no landfills where only MBT residues were disposed of. Even though in Germany and Austria, where MBT technologies have been widely applied, MBT residues have been additionally emplaced on old MSW landfills.

2.3.2 Emission behaviour of MSW landfills

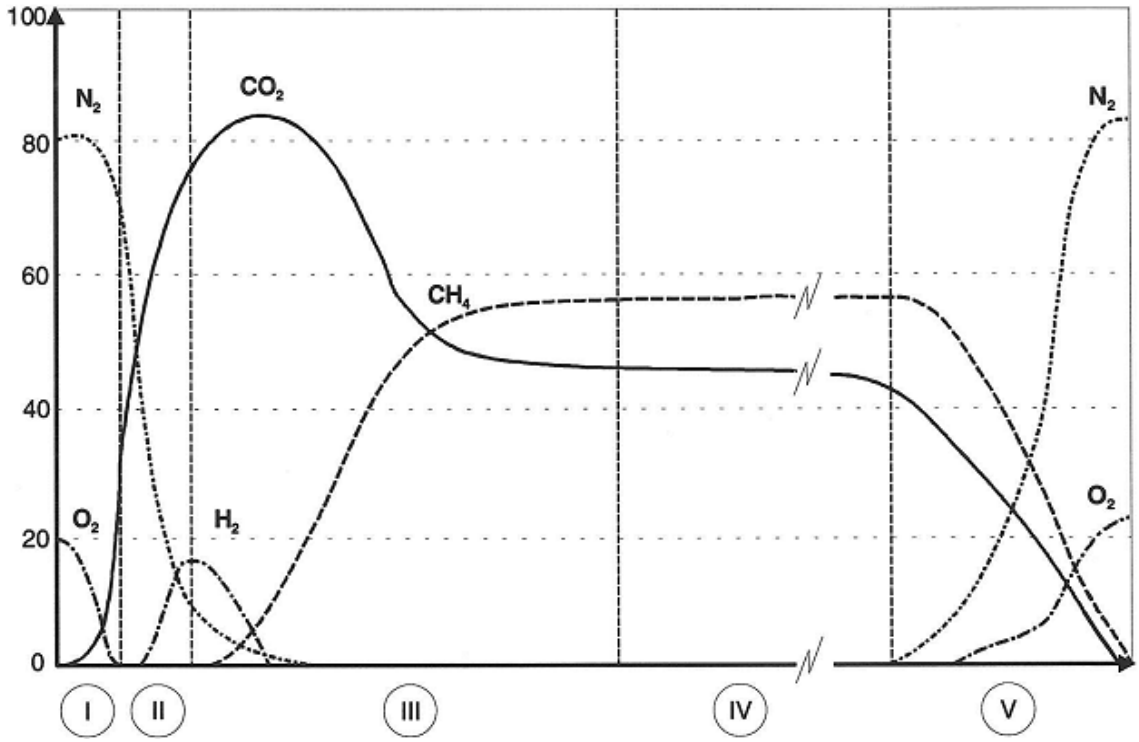
Generally, the environment within a landfill is dependent on many different physical, chemical and biological processes. According to Kjeldsen et al. (2002) and Robinson et al. (2005), it is possible to generalise and identify a set of typical landfill processes, despite large differences in water content, waste composition and management. In landfills with significant amounts of organic waste, biodegradation of organic matters is the dominant process that governs the landfill biogeochemistry. The conversion of organic matters goes through a number of different phases which are comparatively similar between MSW landfills. The number of phases and their names may be varying between publications, but they should express the same principle.

Several landfill investigation studies (Pohland and Harper, 1985) have suggested that the stabilisation of waste goes through five sequential and distinct phases. The rate and characteristics of leachate generated and biogas produced from a landfill vary from one phase to another and reflect the microbially mediated processes taking place in the landfill. According to Pohland et al. (1985), the rate of progress through these phases depends on the physical, chemical, and microbiological conditions developed within the landfill with time.

Due to the fact that landfills have various waste sections or cells with different aging levels, a landfill does not experience a single phase of waste stabilisation but rather many phases of stabilisation occur at the same time.

2.3.2.1 Gas emissions

Landfill gas is a mixture composed of different gases. By volume, landfill gas typically contains 45 – 60 % methane and 40 – 60 % carbon dioxide. Landfill gas also includes small amounts of nitrogen, oxygen, ammonia, sulphides, hydrogen, carbon monoxide, and nonmethane organic compounds (NMOCs) such as trichloroethylene, benzene, and vinyl chloride. The generation of the principal landfill gases is thought to occur in five more or less sequential phases, as illustrated in Figure 2.6 below.



Phase I: Initial adjustment (aerobic); Phase II: Acid formation; Phase III: Initial methanogenic; Phase IV: Stable methanogenic; Phase V: Maturation (second aerobic)

Figure 2.6. Variations in compositions of gas in a landfill cell over time (adapted from: Farquhar and Rovers, 1973; Christensen and Kjeldsen, 1989)

Duration of the individual phases in the production of landfill gas will vary depending on several factors, such as the characteristics of the deposited waste, the distribution of the organic compounds in landfill, the availability of nutrients, the water content of waste, the moisture routing through the fill, the degree of initial compaction and others.

Most landfill gas is produced by bacterial decomposition, which occurs when organic waste is broken down by bacteria naturally present in the waste and in the soil used to cover the landfill.

Landfill gases can be produced when certain wastes, particularly organic compounds, change from a liquid or a solid state into a vapor (known as volatilisation). NMOCs in landfill gas may be the result of volatilisation of certain chemicals disposed of in the landfill.

Landfill gas, including NMOCs, can be also created by the reactions of certain chemicals present in waste. For example, if chlorine bleach and ammonia come in contact with each other within the landfill, a harmful gas is produced.

In phase I, the primary product of decomposition process is carbon dioxide. Decomposition can last for days or months, depending on the availability of oxygen when the waste is disposed of in the landfill. Oxygen levels will vary according to factors such as how loose or compressed the waste was when it was buried. Nitrogen content is high at the beginning of this phase, but slightly declines as the landfill moves through the decomposition phases. Phase I persists until available oxygen is depleted (ATSDR, 2001).

Phase II starts after the oxygen in the landfill has been depleted. The landfill becomes highly acidic. As the acids mix with the moisture present in the landfill, they cause certain nutrients to dissolve, making nitrogen and phosphorus available to the increasingly diverse species of bacteria in the landfill. The gaseous products of phase II are carbon dioxide and hydrogen. If the landfill is disturbed or if oxygen is somehow introduced into the landfill, microbial processes will return to phase I (ATSDR, 2001).

Phase III starts when certain kinds of anaerobic bacteria consume the organic acids produced in Phase II and form acetate, an organic acid. This process causes the landfill to become a more neutral environment in which methane-producing bacteria begin to establish themselves. Methane- and acid-producing bacteria have a symbiotic, or mutually beneficial, relationship. Acid-producing bacteria create compounds for the methanogenic bacteria to consume. Methanogenic bacteria consume the carbon dioxide and acetate, too much of which would be toxic to the acid-producing bacteria (ATSDR, 2001).

Phase IV begins when both the composition and production rates of landfill gas remain relatively constant. Phase IV landfill gas usually contains approximately 45% to 60% methane by volume, 40% to 60% carbon dioxide, and 2% to 9% other gases, such as sulphides. Gas is produced at a stable rate in Phase IV, typically for about 20 years; however, gas will continue to be emitted for 50 years or more after the waste is placed in the landfill (Crawford and Smith, 1985). Gas production might last longer, for example, if

greater amounts of organics are present in the waste, such as at a landfill receiving higher than average amounts of domestic animal waste (ATSDR, 2001).

Regarding phase V, there is a lack of long term scientific data related to this maturation stage. It is generally expected that the landfill mass will eventually evolve towards an aerobic condition as the rate of oxygen diffusion into the waste exceeds the oxygen consumption rate. Other degradation processes, which require an aerobic environment, will then take place. It is believed that the rate at which such a final evolution may progress, or whether it will occur at all, depends on specific landfill conditions such as water content and final cover.

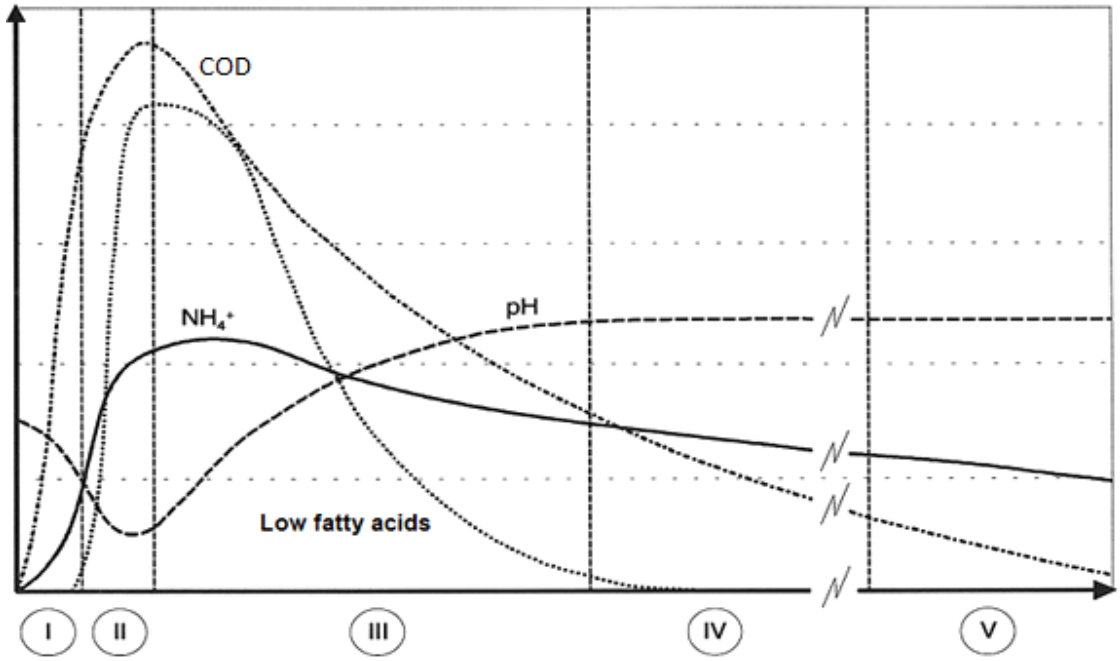
The duration until the ending of the gas production potential in a landfill depends on the degradation rate of organic matter and the gas production rate. In which, the gas production rate is influenced by various factors, such as temperature and water content. It was determined by Reinhardt and Dach (1994) and Reinhardt et al. (1995) that when the water content is less than 15 %, there is no gas formation. The gas formation increases as the water content raises up to 50 %; however, at higher water contents no more increase in gas production rate is detectable.

Landfill gas is known to move from landfills to adjacent areas. The mechanics of gas movement through refuse and soil are extremely complicated. The direction, speed, and distance of landfill gas migration depend on several factors, such as type of landfill cover, natural and man-made pathways, direction and speed of wind, moisture content, groundwater levels, temperature, and barometric and soil gas pressure (ATSDR, 2001).

2.3.2.2 Leachate emissions

According to Rees (1980a), the production of leachate from a landfill site is governed by four principal factors, including: (i) the water content of the waste as being emplaced; (ii) the volume of rainfall allowed to enter the landfill site; (iii) the volume of other liquids added to the waste; and (iv) waste composition and density. These factors not only influence the amount of leachate produced, but also affect chemical composition of the leachate.

Chemical composition of leachate may vary dramatically from one site to another depending on such mentioned factors, as well as the age of landfill, geo-the hydrological properties of the surrounding area, climate conditions, the events preceding the time of sampling, or even the sampling location. However, variations in leachate compositions of landfills generally have identical trends (see Figure 2.7).



Phase I: Initial adjustment (aerobic); Phase II: Acid formation; Phase III: Initial methanogenic; Phase IV: Stable methanogenic; Phase V: Maturation (second aerobic)

Figure 2.7. Variations in leachate compositions in a landfill cell over time (adapted from: Farquhar and Rovers, 1973; Christensen and Kjeldsen, 1989)

Landfill leachate is characterised by a huge number of substances. However, only the relevant chemical parameters for the research are given in the following paragraphs.

pH is a measure of the acidity or basicity of an aqueous solution. The basic principle of electrometric pH measurement is the determination of the activity of the hydrogen ions by potentiometric measurement using a standard hydrogen electrode and reference electrode or a combined electrode. The pH value is an important parameter governing biological processes inside landfills. Leachate from MSW landfills generally has pH values in the range from 4.9 to 9 (Andreottola and Cannas, 1992; Chu et al., 1994; Ehrig, 1980; 1983; and 1988; Johansen and Carlson, 1976; Karstensen, 1989; Krug and Ham, 1997; Lu et al., 1985; Robinson, 1995; and Robinson and Maris, 1979). For a certain landfill, pH values of its leachate depend on the specific phase of the landfill.

Electrical conductivity is a numerical expression of the ability of an aqueous solution to carry an electric current. This ability depends on the presence of ions, their total concentration, mobility, valence, relative concentrations, and on the temperature at the time of the measurement. Solutions of most inorganic acids, bases, and salts are relatively good conductors while molecules of organic compounds that do not dissociate in aqueous solution conduct a current very poorly.

Basically, each conductivity measurement is equivalent to the determination of ion concentrations, with an exception that multi-component systems cannot be analysable with respect to their original components by these measurements in every case. There must be either restrictive conditions or other available variables. For example, acids and alkaline solutions contribute significantly more to the overall conductivity than salts.

Therefore, one possible reason for the variation of the EC in the leachate can be attributed to on the one hand a change in the salt concentration. Possible causes for this are flushing and/or leaching as a result of the leachate recirculation as well as dilution effects due to precipitation and run-off water. The fluctuations can, on the other hand, also be caused by, for example, organic acids, produced or decomposed during the anaerobic metabolic processes.

Ammoniacal nitrogen in leach ate is originated from the nitrogen content of the solid waste, in which its concentration depends on the rate of solubilisation and leaching from the waste (Berge et al., 2005). According to Tchobanoglous et al. (1993), the nitrogen content of MSW is less than 1 % on a wet-weight basis. And it is mostly composed of the proteins contained in food, yard wastes and bio-solids (Burton and Watson-Craik, 1998). As the proteins are hydrolysed and fermented by microorganisms, ammoniacal nitrogen is produced (ammonification process).

In conventional landfills, ammoniacal nitrogen in the leachate primarily presents in form of ammonium because the pH values are normally less than 8.0 (Read et al., 2001; Reinhart et al., 2002). Ammonium is one of the most abundant long-term components in leachate from MSW landfills (Burton and Watson-Craik, 1998; Christensen et al., 2001). It is almost stable or slightly decreased even after centuries as it is not degraded under anaerobic conditions. According to Clement et al. (1997) and Pivato and Gaspari (2005), it is of concern due to its toxicity. Therefore, ammonium has been regarded as a key indicator of groundwater contamination caused by MSW landfill leachate and it is commonly considered during the assessment of pollution risk to groundwater from landfills.

It is well known from literatures that recirculating leachate increases the rate of ammonification, resulting in accumulation of higher levels of ammoniacal nitrogen concentrations than found in conventional landfills, even after the organic fraction of the waste is degraded (Burton and Watson-Craik, 1998; Onay and Pohland, 1998; Barlaz et al., 2002; Price et al., 2003). Hence, ammonium is also the main parameter determining the aftercare period of landfills (Prantl et al. 2006, Ritzkowski et al. 2006). It is also shown from literatures that ammonium concentration in leachate is highly different (from tens to thousands mg/L) from landfill to landfill (Ehrig, 1983; Karadag et al, 2008). This is

common because of the fact that each landfill has its own specificities (e.g. waste composition, weather and climatic conditions...).

Total Kjeldahl nitrogen (TKN) is equal to the sum of ammonium nitrogen and organic nitrogen compounds (TKN = NH_4^+-N + N_{org}). As for landfill leachate, it is important to notice that TKN mostly contains ammonium nitrogen. There are wide variations in TKN and ammonium nitrogen values, reflecting the common fact that each landfill is specified by its own characteristics (Baumgarten and Seyfried, 1996; Ozturk et al., 2003: Tatsi et al., 2003; Frascari et al., 2004).

In consideration of organic compounds, they are generally present in landfill leachate with high concentrations and various species. Organic content in landfill leachate is typically expressed through global parameters such as COD, BOD_5, TOC, and volatile fatty acids (VFA).

The COD is the equivalent amount of oxygen required to chemically oxidise the organic matter contained in a known volume of wastewater using a standard test in which a strong oxidant (potassium dichromate) is used. COD is typically expressed in mg of oxygen/L of wastewater.

The BOD_5 of a wastewater is defined as the amount of oxygen required by aerobic microorganisms to (partially) oxidise the organic matter in a known volume of wastewater in 5 days according to a standard test. BOD is typically expressed in mg of oxygen/L of wastewater.

The TOC of a wastewater is the amount of organic carbon present in a known volume of wastewater as measured in a standard test. TOC is typically expressed in mg of carbon/L of wastewater.

The ratio of BOD_5/COD is frequently used to demonstrate the biodegradability (high BOD_5/COD ratio means high biodegradability and vice versa). The COD/TOC ratio is also occasionally used. It is useful in indicating the degree of oxidation of organic material. A decrease in this ratio reflects a more oxidised state of the organic carbon which becomes less readily available as an energy source for microbial growth (Venkataramani et al., 1974; Chain and DeWalle, 1977). These global parameters and their ratio, however, inadequately provide in details characteristics and species of the organic compounds.

Leachate from young MSW landfills normally has very high COD and BOD_5 values up to 100,000 and 50,000 mg/L, respectively (El-Fadel et al., 2003). Those values may be

even higher or different, depending on the specific MSW landfill. For example, according to Danhamer and Jager (1999) and others (cited in Robinson et al., 2005), COD and BOD_5 in the leachate from landfill/test cells containing untreated mechanically sorted organic residues could reach extremely high values of 172,000 mg/L and 123,000 mg/L, respectively.

In considering the roles of volatile acids in MSW landfills, the literature addressed that VFA and volatile organic acids can affect microorganisms and the degradation processes in two primary ways. First, they have a low ionisation constant (i.e. low pKa) and can readily dissociate, releasing hydrogen ions that cause the pH of the system to decrease and therefore become destabilised. Second, when the acids are non-dissociated (as is typical at low pH levels), the acids are able to penetrate microbial cell membranes, establishing a pH gradient by actively transporting protons out of the cell and reducing the internal cell pH (Aguilar et al., 1995; Zoetemeyer et al., 1982). The decrease in intracellular pH in turn leads to an increased energy demand by the cell to restore pH levels leaving less energy for growth (González et al., 2005; Yamaguchi et al., 1989). These processes lead to reduction in solid waste degradation rate (hinder or delay). High accumulation of VFA can even lead to the interruption of waste degradation.

With respect to sulphate parameter of the leachate, changes in its concentration are used as an auxiliary factor for the interpretation of other parameters and/or processes. According to Rees (1980a), the roles of sulphur compounds in landfill metabolism are intricate. Sulphur is principally present as soluble sulphate and precipitated sulphide with some sulphide in the solution (about 2 mg/L). Sulphur in the forms of SO_4^{2-} and S^{2-} is utilised by microbes for inclusion in the biomass and behaves as an electron acceptor as it is in the form of SO_4^{2-}. Under anaerobic conditions, sulphate reducing bacteria can obtain energy by oxidising simple organic compounds or molecular hydrogen (H_2) while utilising sulphate as an electron acceptor and generate sulphide and alkalinity (Eq. 2-14). Sulphate reducing bacteria can also use sulphites or thiosulphates, and even elementary sulphur as electron acceptors

$$SO_4^{2-} + \text{organic matter} \rightarrow HS^- + H_2O + HCO_3^- \qquad \text{(Eq. 2-14)}$$

The sulphide produced can react with dissolved metals present in the leachate to form metal sulphide precipitates (Eq. 2-15), since the solubilities of most toxic metal sulphides are generally very low (Kim et al., 1999).

$$HS^- + M^{2+} \rightarrow MS(s) + H^+ \qquad \text{(Eq. 2-15)}$$

where M denotes the metal, such as Zn, Cu, Fe, Co, or Ni.

The metal precipitation reaction releases protons, thus decreasing pH value of the leachate. HCO_3^- alkalinity produced in the sulphidogenic oxidation of organic matters (see Eq. 2.14) neutralise the acidity of the leachate (Eq. 2-16).

$$HCO_3^- + H^+ \leftrightarrow H_2O + CO_2 \qquad \text{(Eq. 2-16)}$$

Sulphate reducing bacteria metabolism generates sulphide which can inhibit microorganism activities. According to Percheron et al. (1997), Lens et al. (1998), and Weijma et al. (2002), the free soluble form of sulphide (i.e. H_2S) can seep into cell membranes and structure cross-links between polypeptide chains, thus changing cell proteins. While H_2S appears to be correlated to methanogenic bacteria, there has been indicated that dissolved sulphide correlates to inhibition of sulphate reducing bacteria (Hilton and Oleszkiewicz, 1988; Parkin et al., 1991; Visser et al., 1996).

The introduction of air into a landfill and/or its leachate can lead to the aerobic oxidation of reduced sulphur compounds (Eqs. 2-17, 2-18, and 2-19).

$$HS^- + 2\ O_2 \rightarrow SO_4^{2-} + H^+ \qquad \text{(Eq. 2-17)}$$

$$H_2S + 2\ O_2 \rightarrow SO_4^{2-} + 2\ H^+ \qquad \text{(Eq. 2-18)}$$

$$H_2S + ½\ O_2 \rightarrow S + H_2O + \text{energy} \qquad \text{(Eq. 2-19)}$$

When nitrate is present in the system, denitrifying ammonium oxidation (DEAMOX) process may also occur (Eq. 2-20).

$$4\ NO_3^- + S^{2-} \rightarrow 4\ NO_2^- + SO_4^{2-} \qquad \text{(Eq. 2-20)}$$

Table 2.1 shows the variations in concentration of different leachate parameters against landfill age (after starting a new landfill or landfill cell) and limit values for the discharge of treated leachate according to the German standands. These data were obtained from 76 landfills in Germany.

Table 2.1. Average concentration of selected leachate parameters versus landfill age (adapted from Krümpelbeck and Ehrig, 1999) compared to the German discharge limits (AbwW, 1996)

Parameter	**Unit**	**1 - 5 years**	**6 - 10 years**	**11 - 20 years**	**21 - 30 years**	**German discharge limits**
pH	-	7.3	7.5	7.6	7.7	-
EC	mS/cm	9.28	12.16	10.61	12.93	-
TOC	mg/L	1,235	845	520	475	10[a]
COD	mg/L	3,810	2,485 – 3,255	1,585 – 1,830	1,160 – 1,225	200
BOD_5	mg/L	2,285	800 – 1,210	275 – 465	185 – 290	20
NH_4-N	mg/L	405	600	555	445	70[b]
NO_3-N	mg/L	3.6	7.6	11.7	9.2	
NO_2-N	mg/L	0.064	0.63	0.54	0.84	2
SO_4^{2-}	mg/L	98	146	93	83	1[c]

[a]: Total hydrocarbons; [b]: Sum of NH_4-N + NO_3-N + NO_2-N; [c]: Sulphide

It can be clearly seen from Table 2.1 that most of the selected leachate parameters have concentrations which are higher than the German discharge limits. Nitrite nitrogen is low because there is normally no pathway for its formation under anaerobic condition of MSW landfills.

Regarding a certain landfill, leachate concentration may greatly change with operational time. Table 2.2 shows the quality of leachates during the acidic and methanogenic phases of an MSW landfill.

Table 2.2. Typical concentration of selected leachate constituents of an MSW landfill (adapted from Ehrig, 1990)

Parameter	Unit	Acidic phase		Methanogenic phase	
		Mean	***Range***	***Mean***	***Range***
pH	-	6.1	4.5 – 7.5	8.0	7.5 – 9.0
COD	mg/L	22,000	6,000 – 60,000	3,000	500 – 4,500
BOD_5	mg/L	13,000	4,000 – 40,000	180	20 – 550
TOC	mg/L	7,000	1,500 – 23,000	1,300	200 – 5,000
SO_4^{2-}	mg/L	500	70 – 1,750	80	10 – 420

In another survey, constituents in the leachate during the acidic, intermediate, and methanogenic phases of an MSW landfill are given in Table 2.3.

Table 2.3. Constituents in leachates from an MSW landfill (adapted from Kruse, 1994)

Parameter	Unit	Acidic phase		Intermediate phase		Methanogenic phase	
		Mean	***Range***	***Mean***	***Range***	***Mean***	***Range***
pH	-	7.4	6.2 – 7.8	7.5	6.7 – 8.3	7.6	7.0 – 8.3
COD	mg/L	9,500	950 – 40,000	3,400	700 – 28,000	2,500	460 – 8,300
BOD_5	mg/L	6,300	600 – 27,000	1,200	200 – 10,000	230	20 – 700
DOC	mg/L	2,600	350 – 12,000	880	300 – 1,500	660	150 – 1,600
SO_4^{2-}	mg/L	200	35 – 925	90	20 – 230	240	25 – 2,500

DOC: dissolved organic carbon

The data from Tables 2.3 and 2.4 show significant differences between the two investigations concerning the organic parameters. The results from Ehrig (1990) indicate that leachate concentrations of COD, BOD_5, and TOC in the older landfills are higher than those determined by Kruse (1994) some ten years later. Such differences can be explained by certain developments in the waste landfilling technology, in which waste compaction in many younger landfills is implemented in thin layers. Additionally, the waste composition may have changed towards less biodegrable waste being landfilled (Stegmann et al. 2005).

While variations in organic parameters change with landfill age, nitrogen parameters are independent from the sequential phases of landfills (see Table 2.4).

Table 2.4. Nitrogen parameters in leachates from MSW landfill (adapted from Ehrig, 1990 and Kruse, 1994)

Parameter	**Unit**	**According to Ehrig, 1990**		**According to Kruse, 1994**	
		Mean	***Range***	***Mean***	***Range***
TKN	mg/L	1,350	40 – 3,425	920	250 – 2,000
NH_4-N	mg/L	750	30 – 3,000	740	17 – 1,650
NO_3-N	mg/L	3	0.1 – 50	N/A	N/A
NO_2-N	mg/L	0.5	0 – 25	N/A	N/A

N/A: not available

Although there are large variations at individual MSW landfills, a decreasing tendency of concentrations can be typically observed for organic carbon compounds in leachate over landfill age. Whereas, concentrations of ammonium nitrogen are still high, which fluctuate and/or slightly decrease over a long period of time.

2.3.3 Emission behaviour of MBT landfills

In MBT landfills the waste degradation phases undergo significant changes in comparison to those occurring in MSW landfills. Accroding to Bockreis and Stainberg (2005), Cappai et al. (2005), and De Gioannis et al. (2009), the major change is the reduction of stages prior to the methanogenic phase. This reduction occurs because aerobic phase has already finished during the biological stage of MBT process (Bockreis

et al. 2003). The degree of biological stage can strongly affect the reduction of these phases. De Gioannis et al. (2009) stated that the reduction in the duration of non-methanogenic phases for a material subjected to aerobic treatment of 8 and 15 weeks is 67 and 82 %, respectively, compared to untreated waste. However, long-term emission behaviours from pure MBT landfills (landfills contain only MBT residues) are either very limited or unavailable, because of the fact that MBT residues typically are deposited together with variable proportions of untreated MSW, mechanically sorted organic residues, commercial and industrial wastes (Robinson et al., 2005).

For this reason, different authors have studied such behaviours using landfill simulation reactors (LSR) in laboratories. Their simulation experiments can be divided into two groups. The first group involves the studies of so-called "classical" LSR, which are characterised by a relatively low overall density (up to maximum of 0.5 kg DM/dm^3) and high rate of leachate exchange. The second group is carried out in LSR which have "more realistic" conditions.

Due to low permeability of the MBT material, on one hand surface run-off occurs and on the other hand the high saturation of the top level of the waste layer leads to a squeezing of water and soaking of the waste, which makes a passing over of the surface impossible (Münnich et al., 2006a). Therefore, MBT landfills expect a much lower rainfall infiltration and leachate generation. For these reasons, the treated wastes (MBT residues) were compacted to high density (up to approximately 0.9 kg DM/dm^3) in LSR. By doing so, the influence of loading density and water content on emission behaviour can be evaluated (Leikam, 2002).

2.3.3.1 Gas emissions

As a consequence of shortening degradation phases by MBT process, biogas production is considerably reduced in MBT residues after the disposal. The reduction of biogas production depends on the duration, degree and the type of biological treatment stage. In general, the longer and more intense the biological treatment is applied, the lower the gas generation potential is expected. MBT reduces the landfill gas emission potential by 90 % (Höring et al., 1999) or more compared with that from untreated MSW (Scheelhaase and Bidlingmaier, 1997; Doedens et al., 2000; Zach et al., 2000; Lornage et al., 2007). De Gioannis et al. (2009) observed that the total volume of gas generated by waste decreased by 80% after an intensive aerobic treatment of 8 weeks, while for a treatment of 15 weeks the decrease was 91 %. Sormunen et al. (2008) stated a reduction in gas production potential of 80 % in a composting system that combines aerated pilot tunnels (2 - 3 weeks) and passively aerated pile composting outdoors (6 - 14 months) for mechanically produced residues (residues after mechanical treatment). Such reduction was reported to

be more than 90 % by Lornage et al. (2007) after 25 weeks of biological treatment by means of forced aeration. Due to a significant reduction in landfill gas production achieved by an adequate MBT process, a landfill gas collection and extraction system may be no longer necessary if the deposited residues have been stabilised sufficiently. Passive filter systems and/or methane oxidation layer may be needed under these conditions.

Scheelhaase et al. (1999) stated a decrease in gas production with increasing residue density. Studies from Dach (1998) and Scheelhaase et al. (1999) also largely affirmed that the gas permeability decreases by increasing residue densities and decreasing gas pore space. Therefore, the gas movement in MBT landfills is lower compared to that in MSW ones.

2.3.3.2 Leachate emissions

In considering the amount of leachate generated from MBT residues, data from literatures showed inconsistent results. Kuehle-Weidemeier and Doedens (2003) addressed that the amount of leachate from MBT residues is very high compared with MSW, despite the low permeability of the pre-treated residues. On the contrary, Münnich et al. (2005 and 2009) pointed out that the increased density of MBT residues after their emplacement on landfills results in the reduction of leachate generation. Also according to Müller and Bulson (2005), MBT residues can be compacted to a high density (> 1 Mg/m^3 dry waste) and has a very low hydraulic conductivity ($5*10^{-7}$ - 10^{-10} m/s). Such mechanical properties will reduce leachate emission rates and will allow the use of smaller leachate treatment systems.

As for the quality of leachate released from MBT residues, Höring et al. (1998) and Scheelhaase et al. (1999) stated that, basically, it can be expected the more intensive and longer MSW waste is treated, the less mobilised substances in the output residues, therefore in the leachate produced are achieved. Höring et al. (1998) also stated that the leachate emission potential from biologically stabilised wastes is reduced by about 95 % in terms of TOC (comparing to the acid phase of conventional landfills) and by about 40 - 90 % regarding nitrogen parameters. In another study, Leikam and Stegmann (1999) addressed that because of MBT process, BOD_5, COD and total nitrogen content in the leachate generated from MBT residue decreased by about 90 % in comparison with those from MSW.

In LSR studies with highly compacted wastes having $RI_4 \leq 5$ mg O_2/g DM, Dach (1998) and Scheelhaase et al. (1999) obtained data which show significantly higher

concentrations of TOC compared with the investigation of Höring et al. (1998); however, there was only little higher concentrations of nitrogen parameters (see Table 2.5).

Table 2.5. Leachate of MBT waste in the simulation experiment compared to leachate of MSW landfills in the methane phase

Author		**Type of investigation**	**TOC (mg/L)**	**TN (mg/L)**	**NH_4-N (mg/L)**
MBT	Höring et al. (1998)	LSR test - low compacted	< 600	200 – 400	N/A
	Dach (1998)	LSR test - high compacted	637 – 2250	243 – 514	188 – 266
	Schneelhaase et al. (1999)	LSR - high compacted	500 – 2000	400 – 600 (TKN)	N/A
Untreated MSW	Ehrig (1989)	Evaluation of landfill data	1000 (COD: 3000)	1250	750
	Kruse (1994)	Data analysis of 33 landfills	833 (COD: 2500)	920 (TKN)	740
	Krümpelbeck and Ehrig (1999)	Data analysis of 70 landfills (11 - 20 years old)	520	N/A	555

N/A: not available

Dach (1998) assumed that due to the low volumes and long residence times of leachate in the reactor, a chemical equilibrium between leachate and solid phase is established so that the measured concentrations should be similar to those of real leachate from a highly compacted landfill. Overall, the concentrations of nitrogen parameters (ammoniacal nitrogen and TN or TKN) in the simulation studies for treated waste in both cases of low and high installation density are reduced by about 50 % in compared to those of untreated MSW in the methanogenic phase.

As for field studies, Table 2.6 shows the data for leachate from two MBT landfills in Germany.

Table 2.6. Leachate quality from MBT landfills (adapted from Doedens et al., 2000; Hertel et al., 2001)

Parameters	**Unit**	**MBT Lüneburg - first 2 years**	**MBT Erbenschwang - first 3 years (1999 - 2001)**	
			Mean	***Range***
RI_4	mgO_2/g DM	2.5 – 12	*N/A*	10 – 34
pH	-	7.5	8.7	8.4 – 9.5
EC	mS/cm	*N/A*	9.5	4.12 – 15.2
TOC	mg/L	300 – 950	1,161	294 – 1,760
COD	mg/L	700 – 2,500	3,634	860 – 6,260
BOD_5	mg/L	1 – 55	65	3 – 322
NH_4-N	mg/L	0 – 27	*N/A*	*N/A*
NO_3-N	mg/L	15 – 66	436	23 – 911
NO_2-N	mg/L	0.1 – 1.7	*N/A*	*N/A*
TN	mg/L	35 – 140	*N/A*	*N/A*
TKN	mg/L	10 – 37	291	70 – 428
SO_4^{2-}	mg/L	*N/A*	378	252 – 676

N/A: not available

The data in Table 2.6 clearly show that ammonium and total nitrogen concentrations in the leachate from Lüneburg MBT landfill are critical low; meanwhile, nitrate nitrogen is relatively high. In case of Erbenschwang MBT landfill, nitrate nitrogen in the leachate is extremely high, whereas data for ammonium and total nitrogen are not available. High concentrations of nitrate nitrogen in the leachate imply that nitrification process has taken place either before (during biological treatment at the MBT plants) or after the emplacement of MBT residues on those landfills.

Additionally, the MBT materials in both landfills have average RI_4 values higher than the allowed value of 5 mgO_2/g DM, especially in case of Erbenschwang MBT landfill. This means that the organic matters in the MBT residue are higher than expected.

Taking into account the emission behaviour of both MSW and MBT landfill types, the long persistence of ammonium in leachate is emerged as a critical concern. Although MBT processes have successfully brought to a significant reduction of carbonaceous organics and nitrogen in the MBT output residues in comparison to the MSW input materials, ammoniacal nitrogen still exists at relatively high concentrations.

2.4 Techniques for in-situ stabilisation of organic matter in landfills

One of the main goals of waste management is the development of so-called "sustainable landfill" concept. "Sustainable landfill" can be understood as a landfill where the waste mass is nearly in a stable state, meaning that the remaining conversion processes are low and emission release is below the acceptable level. However, it takes very long time for landfills to be sustainable if no management strategies are applied. Both gas and leachate emissions could last for hundreds of years, of which leachate emission is of more critical concern due to its compositions causing several hazards to the environment. Therefore, treatment of the leachate plays a key role in the whole waste management process.

Wherever leachate is collected, a discharge option must be provided. It is often that leachate requires treatment before final discharge to the environment in most case (Johannessen, 1999).

Treatment of leachate can be either locally on-site (in-situ) or off-site (ex-situ). Local treatment can be physical-chemical, biological or a combination of both. Meanwhile, the latter means that leachate is transferred to another place (most commonly in Municipal Waste Water Treatment Plants), where it will be mixed with other wastewaters. Ex-situ treatment typically involves biological processes.

The main components to be treated in MSW leachate are organic matter, ammonia, and chlorides. The type and degree of treatment may greatly vary, depending on standards for discharge or the vulnerability of the receiving water (where such standards do not exist), climatic conditions, and the quality and quantity of leachate generated. Therefore, a combination of different methods may be necessary (Johannessen, 1999).

Using the leachate for irrigation or recirculating it back to the landfill is other options. In such case, the landfill can be referred to as bioreactor landfill (Reinhart and Al-Yousfi, 1996; Cheng and Chu, 2007).

According to Cossu (2005), a landfill is considered to be sustainable if it reaches an equilibrium state with the environment within a period of 30 - 40 years. As considering whether a landfill becomes sustainable, gas and in particular leachate emission are the

key parameter. It is necessary to enhance waste stabilisation in order to reduce the time required for leachate treatment. According to Kylefors (1997), waste stabilisation can be hindered by different factors, including lack of moisture, lack or poor distribution of nutrients, accumulation of degradation products, and small contact area of waste, water and microorganism. Therefore, attempts in avoiding or limiting such hindering factors should enhance the stabilisation process. In another respect, stabilisation can be achieved via: (i) pre-treating the waste by size reduction, mixing and pre-composting; and (ii) using flow systems to influence the environmental conditions within the landfill (ARRPET, 2004).

Figure 2.8 summarises a list of possible technologies and thereof combinations to achieve a sustainable landfill.

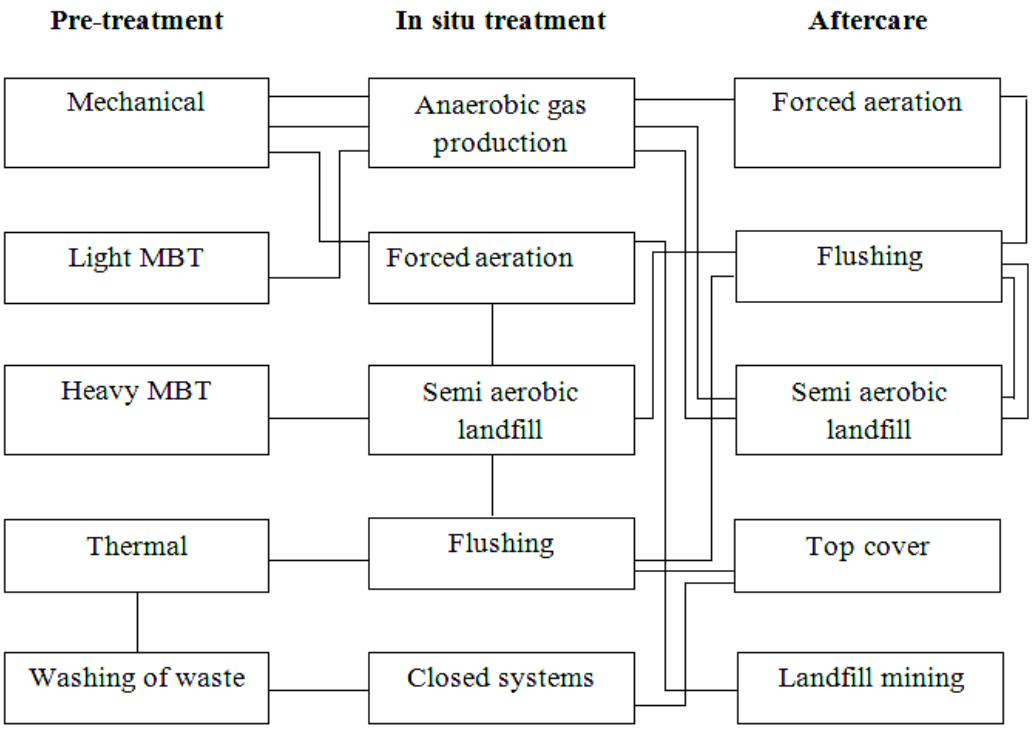


Figure 2.8 Combination of technologies for approaches to "sustainable landfill" (adapted from Cossu, 2005)

It can be seen from Figure 2.8 that pre-treatment is a prerequisite requirement, in which the incorporation of MBT technologies in MSW management is involved. Also in regards of such combination techniques, aeration and flushing measures are dominating others.

2.4.1 Flushing measure

Flushing measure principally targets at two main goals. Firstly, it provides suitable water content for the landfill body, in which microbial bacteria are facilitated to grow and speed

up biological conversions. The second goal is that various organic and inorganic compounds can be flushed or leached out of the solid waste. Such outcomes could contribute to improve the stabilisation of the landfilled waste.

In order for the flushing process to occur, the water content of the waste must be higher than its water holding capacity. The effectiveness of flushing process will depend on porosity and the pore size distribution of the waste. The problem in the landfill body is areas with preferential flow, while in other areas nearly no flow occurs. The effectiveness is then reduced considerably. In case of MBT landfills, preferential flow paths are reduced because of the higher homogeneity of deposited MBT materials, but they still exist.

Also regarding MBT landfills, it is more difficult to introduce large volumes of liquids into the landfill body because of the usually lower permeability compared with that of MSW ones.

Flushing can be done by either flushing of landfilled waste with water or by recirculation of leachate back into the landfill body. Flushing of waste with water can reduce concentrations of several pollutants in leachate very fast, but actually requires a huge volume of water to pass throughout the waste mass. Leachate recirculation can also provide opportunities for pollutant reductions, but at lower extent in comparison to flushing of waste with water. Both methods can reduce the time required for biological stabilisation of the readily biodegradable leachate constituents and increases the rate of leachate bio-stabilisation. Kylefors (1997) stated that leachate recirculation enhances landfill stabilisation by removing the waste products after degradation from the liquid phase and allowing addition and distribution of microorganisms and nutrients within the landfill body. According to Pohland and Harper (1985), leachate recirculation measure can reduce the stabilisation time from 15 - 20 years to 2 - 3 years.

However, leachate recirculation and flushing of waste with water in particular both could increase risk related to the existence of preferential flows within the landfill body, leading to the local flushing or in worst case may deteriorate the physical stability of landfill. Principally, the higher the volume of water in the landfill body, the lower physical stability of the landfill.

Laboratory tests performed by Pohland (1972 and 1975), Leckie et al. (1975 and 1979) and Pohland et al. (1990) showed that leachate recirculation leads to rapid decline in concentrations of COD, BOD_5, TOC, volatile fatty acids (VFA), phosphate, and ammonium. Whereas, a full-scale study on leachate recirculation by Robinson and Maris (1985) indicated that the recirculated leachate is still high in concentrations of ammonium

and COD. The results from the full-scale study seem to be more coherent because ammonium is accumulated by the recirculation and there are no conversion pathways for it under anaerobic conditions in conventional landfills.

2.4.2 Aeration

Aeration simply refers to the introduction of air or oxygen into landfilled refuse, in which air or oxygen addition can be done by means of high- or low-pressure pumping. Aeration can be either intermittent, continuous or a combination of such regimes.

The high-pressure aeration was first applied in Austria, in which ambient air enriched with oxygen was used to free up methane out of an old MSW landfill (Dörrie et al., 1998). The technique has also been successfully used for the reduction of odours and methane concentrations during mining of old landfill sites (Heyer et al., 2001; Hogland et al., 2004; Stegmann and Ritzkowski, 2007).

In addition to the application in the landfill mining domain, in-situ aeration is a promising treatment technique for the stabilisation of the landfilled refuse and for the reduction of leachate treatment needs (Read et al., 2001; Ritzkowski et al., 2006; Rich et al., 2008). Operating the landfill aerobically has shown additional advantages, including increased settlement, decreased metal mobility, lower methane control costs, and reduced environmental liability (Environmental Control System, Inc., 1999; Read et al., 2001).

During aerobic degradation of MSW, biodegradable materials are mostly converted to carbon dioxide and water. In theory no methane is produced during aerobic degradation. In practice an absolute aerobic condition does not seem to exist in landfills, regardless of initial adjustment (aerobic) phase or forced aeration. Certain areas of the solid waste still have anaerobic condition. Therefore, little (if any) methane is produced and volatile organic acid production is decreased. Moreover, nitrogen transformation and/or removal processes (nitrification, ammonia air stripping or volatilisation) are favoured (Berge et al., 2005).

In-situ field-scale tests have been conducted to investigate the effects of low-pressure aeration on MSW waste stabilisation and found that in-situ aeration has a significant potential for reducing landfill emissions and also the costs necessary in the aftercare stage (Ritzkowski et al., 2006).

2.5 Summary and problem statement

Despite the fact that landfilling of MSW is world-wide considered to be primarily a waste disposal operation (Rees, 1980a; Tchobanoglous et al., 1993; Erses et al., 2008), many

countries in the mean time, however, are facing challenges when their landfill sites are reaching full capacities and especially when environmental, health and safety concerns are prominent (Ali, 1999; Cheremisinoff, 2003).

One of the most environmental concerns is caused by the emissions of landfill gas and leachate, of which leachate emissions definitely pose several prolonged hazards. After landfilling, the treatment of generated leachate should be the most important task regarding the overall management of the landfill. However, landfill management is a complex task due to the highly variable nature of landfilled waste type, design and age of the landfill, operational expenditures as well as climatic and seasonal conditions.

Leachate treatment depends on several aspects, such as the quality and quantity of the leachate input, discharge limits or removal efficiency requirements, quantity of residual products and their management, site location, and economics.

In aspect of leachate treatment, high concentrations of organic compounds, ammonium, and heavy metals are generally the key factors. Therefore, treatment techniques typically aim at diminishing these components as much as possible at reasonable costs.

In several developed countries where direct landfilling of non-treated MSW has already been forbidden, but a lot of old closed MSW landfills have still been posing threats on environment due to their leachate emissions. Meanwhile, in most developing countries MBT technologies have not been, or very seldom applied, MSW management still ordinarily relies on the direct disposal of the waste in landfills. Therefore, leachate emissions are in extremely high loads of organic substances as well as ammonium concentrations.

The incorporation of MBT technologies in MSW management has recently applied in many developed countries. It has been increasingly popular in the member states of the EU, of which Germany has possessed the major portion. By comparing with conventional landfilling, this new measure has brought about many advantages, including: reduction in quantity of residues being landfilled and also in volume and strength of leachate and gas produced; avoidance of clogging of leachate drainage systems; and shorter timescale to waste stabilisation and lower costs for landfill aftercare (Leikam and Stegmann, 1996; Robinson et al., 2005; Münnich et al., 2006b).

The main objective of MBT is to reduce organic carbon compounds in the output material to be deposited and therefore also in the leachates produced in comparison to those of fresh MSW (Leikam and Stegmann, 1999; Robinson et al., 2005; Siddiqui et al., 2012). The small number of leachate data from MBT landfills shows that TOC concentrations

are considerably reduced, while the ammonium concentrations still remain at a relatively high level compared with the discharge limits (Münnich et al., 2011). Therefore, a complementary technique is needed to reduce also these emissions.

As mentioned in section 2.4.1 that the laboratory and full-scale studies involving recirculation of leachate for MSW have engendered the results for organic carbon compounds and ammonium concentrations in leachate, which are not consistent. Therefore, an investigation on whether recirculation of leachate could bring to positive effects on leachate quality is still of meaningfulness.

In literature, leachate recirculation has been successfully utilised either solitarily or in combination with aeration, however these techniques were only regarded to MSW taken from old MSW landfills (at lab-scale) or the old MSW landfills themselves (at field scale). Moreover, regarding the combined technique it has been not clearly shown whether intermittent or continuous operation giving better effectiveness.

From what have been discussed above, the following statements need to be clarified whether:

- Recirculation of leachate can reduce concentrations of organic substance and ammonium in leachate from fresh MSW and MBT residues;
- Combination of leachate recirculation and aeration can be appropriately used for MBT residues which are much stabilised in comparison with MSW;
- Such combination technique can give a positive effect on fresh MSW; and
- Intermittent or continuous aeration regimes are more effective.

CHAPTER 3: MATERIALS AND METHODS

3.1 Introduction

The investigations were performed in laboratory scale. Series of lysimeters with different dimensions and defined boundary conditions have been used to simulate different types of bioreactor landfills.

The whole experimental tests have been divided into three stages. In the first stage (phase 1), four 141-L lysimeters were run under leachate recirculation to simulate anaerobic bioreactor landfills. Amongst four lysimeters, two contained MBT residue (lysimeters 1 and 2) and two others (lysimeters 3 and 4) were loaded with fresh MSW. The aim of phase 1 is to investigate the effect of leachate recirculation on the quality of generated leachates.

The second stage (phase 2) involves a set of four small 1.34-L lysimeters all containing MBT residue. Of which, two lysimeters were operated under only leachate recirculation and two others under the combination of leachate recirculation and intermittent aeration. The purpose of phase 2 is to investigate whether such combination technique give positive results in improving the quality of leachates generated. It is also a preliminary step prior to investigate such effects as well as some boundary conditions at a larger scale in the third phase.

The operation of phase 1 and phase 2 was implemented in room temperature condition of about 20 °C.

In the third stage (phase 3), a set of six 141-L lysimeters was used. Lysimeters 1 and 2 were loaded with MBT residue and four others with MSW. Amongst four MSW lysimeters, two lysimeters 3 and 4 from phase 1 were re-used; however, the lysimeter 3 was opened to install an aeration tube, whereas the lysimeter 4 was kept unchanged. The MSW inside these two lysimeters has undergone an anaerobic duration of 349 days. Two remaining lysimeters 5 and 6 were loaded with fresh MSW and were operated under leachate recirculation solely for a certain time. As leachate parameters reach relatively stable levels, the combination regime of leachate recirculation and intermittent aeration were applied. Four lysimeters 1, 2, 3, and 4 were operated under room temperature condition of about 20 °C, whereas two lysimeters 5 and 6 were under warm condition of about 30 °C. Five lysimeters 1, 2, 3, 5, and 6 were installed aeration system and run under different boundary conditions of aeration. Whenever in-situ aeration was applied, the lysimeter was used to simulate hybrid bioreactors.

Phase 3 aims to investigate the influences of different in-situ aeration regime in combination with leachate recirculation on quality of the leachates generated from MBT residue, as well as 1-year old MSW and fresh MSW under the defined boundary conditions.

The brief summary of such phases and the relating lysimeters is given in Table 3.1 below.

Table 3.1. Summary of three experimental phases

Phase	Number of lysimeter	Lysimeter dimension	Loading material	Temperature condition	Test duration
1	4	d = 41 cm H = 107 cm V = 141 L	- LSR 1 and 2: MBT residue - LSR 3 and 4: Fresh MSW	about 20 °C	295 days 250 days
2	4	d = 10 cm H = 17 cm V = 1.34 L	MBT residue	about 20 °C	85 days
3	6	d = 41 cm H = 107 cm V = 141 L	- LSR 1 and 2: MBT residue - LSR 3 and 4: One-year old MSW	about 20 °C	413 days
			- LSR 5 and 6: Fresh MSW	about 30 °C	310 days

d: diameter; H: height; V: volume

3.2 Materials and analytical methods

Two types of material (fresh MSW and MBT residue) were used throughout the whole research programme for the investigation of different parameters. The materials were sampled at the MBT plant in Göttingen, Germany. In this plant anaerobic fermentation is coupled with aerobic stabilisation. The fresh MSW is the input material for the fermentation process. The MBT residue is the output which is in the range of the allowed values and concentrations defined in the German requirements for the emplacement on landfills (DepV, 2009).

The materials used for each experimental phase were sampled at a specific time. And for each group of materials, they are comparatively similar regarding their compositions and characteristics.

After sampling, the waste was kept in plastic containers with closed caps, marked and stored in climatic cooling chambers.

3.2.1 Characteristics of input and output materials

Regarding each type of the materials, some certain portions have been representatively collected and prepared to determine the characteristics of the materials before loading into (or unloading from) the lysimeters. The input and output materials are characterised by analyses of the solid phase and its eluate. The solid phase was examined for water content and biodegradability which is expressed via respiratory activity index after 4 days (RI_4). The eluate was investigated for nitrogen and organic carbon parameters.

3.2.1.1 Water content

Water content is determined according to the German standard DIN ISO 11465: 1996-12. The wet waste sample is weighed and then dried at 105 ^{o}C for at least 24 hours to constant weight. The dried waste sample is weighed again and the water content is calculated based on the difference between the wet and dried waste masses in comparison to the mass of the wet waste sample.

For each type of material, at least three wet samples are determined for their water contents. The final water content of the wet waste is the average value calculated from the individual values.

3.2.1.2 RI_4

RI_4 is determined in accordance with the DIN ISO 16072: 2002 using the WTW OxiTop®Control system. The principle is based on the measurement of pressure changes in a closed vessel. The closed vessel containing the waste sample to be analysed is kept in an incubation chamber at 20 °C. Microorganisms in the waste sample consume oxygen and form carbon dioxide. The carbon dioxide produced is absorbed by a NaOH solution. In the closed system, the change in pressure is proportional to the amount of oxygen being consumed.

3.2.1.3 Eluate characteristics

Eluate is prepared by the extraction of the dried waste with deionised water (Liquid/Solid ratio = 10) in 2L-plastic bottles. The bottles are shaked for 24 hours using a shaking machine. After stopping, the upper part is separated and then filtered through a 0.45 μm pressure filtration membrane. The procedure complies with DIN 38414-S4.

The filtrate is evaluated for nitrogen (NH_4-N, NO_3-N, NO_2-N) and organics (TOC, COD) parameters. Details of those analyses are given in sections 3.2.2.2 and 3.2.2.3.

3.2.2 Characteristics of the leachate and gas during experimental operation

Leachate has normally been sampled twice a week. Right after the sampling, one part of the leachate is measured for pH and EC values. The other part is prepared and/or preserved for the determination of nitrogen and organics parameters afterwards. Leachate samples are generally being analysed in the same day of sampling. If not possible, the samples are preserved in the freeze at 4 ^oC and in accordance with the preserved precautions.

Gas analysis has normally been done on the basis of one week; however, it is also dependent on whether there are any gases in the gas collection bag.

3.2.2.1 pH and EC values

pH and EC values were measured using equipments WTW pH 315i WTW and WTW Cond 315i in accordance with DIN 38404-C5 and -C8, respectively.

3.2.2.2 Nitrogen parameters

Nitrogen parameters were characterised by the analyses of the leachate and eluate for ammonium, nitrate, nitrite, and TKN or TN concentrations.

Regarding the determination of ammonium, nitrate and nitrite, the leachate had to be filtered through a 598½ filter paper. The filtrate was then used for the analysis. The filtrate might have needed further dilutions depending on whether the concentration of the measured parameters was in the range of the analytical method being used. Ammonium, nitrate, and nitrite nitrogen were determined using corresponding Hach Lange cuvette test kits in combination with Spectrophotometer Dr 2800.

Determination of TKN or TN does not require the filtration step, but the leachate samples need to be homogenised before digestion procedures. The homogenised liquid might also need further dilutions similar to those applied to ammonium, nitrate and nitrite determinations. In

phase 1 and the first half of phase 2, the determination of TN (using Hach Lange cuvette test kits) was not available. TKN was determined by Method 8075 which involves the digestion of the sample with the mixture of concentrated H_2SO_4 and H_2O_2 at 440 ^oC. The receiving digested solution is then photometrically determined with Spectrophotometer Dr 2800. High temperature in the digestion process is prone to pose errors due to the loss of sample.

The principle for the determination of nitrogen parameters is summarised in Table 3.2.

Table 3.2. Principle for determination of nitrogen parameters

Parameter	**Principle**	**Standard**
Ammonium	Ammonium ions react at pH 12.6 with hypochlorite ions and salicylate ions in the presence of sodium nitroprusside as a catalyst to form indophenol blue.	ISO 7150-1 or DIN 38406 E5-1
Nitrate	Nitrate ions in solutions containing sulphuric and phosphoric acids react with 2,6-dimethylphenol to form 4-nitro-2,6-dimethylphenol.	ISO 7890-1-2-1986 or DIN 38405 D9-2
Nitrite	Nitrites react with primary aromatic amines in acidic solution to form diazonium salts. These combine with aromatic compounds that contain an amoni group or a hydroxyl group to form intensively coloured azo dyes.	EN ISO 26777 or DIN 38405 D10
Total nitrogen (TN)	Inorganically and organically bonded nitrogen is oxidised to nitrate by digestion with peroxo-disulphate. The nitrate ions react with 2, 6-dimethylphenol in a solution of sulphuric and phosphoric acid to form a nitrophenol.	EN ISO 11905-1 or DIN 38409 H-36
Total Kjeldahl nitrogen (TKN)	TKN refers to the combination of ammonia and organic nitrogen. However, only the organic nitrogen compounds appearing as organically bound nitrogen in the trinegative state are determined. Nitrogen in this form is converted into ammonium salts by the action of sulphuric acid and hydrogen peroxide. The ammonium is then analysed by a modified Nessler method test. Test results are measured at 460 nm.	Method 8075 - Nessler method[(a)]

[(a)]: Adapted from Hach et al., 1985 and Hach et al., 1987

3.2.2.3 Organic parameters

Organic parameters in the leachate were investigated by determination of COD, TOC and fatty acids using the corresponding Hach Lange cuvette test kits and Spectrophotometer Dr 2800. Fatty acids data are available for phase 3 only. Determination of BOD_5 was carried out by dilution method. However, the sensor for the measurement of dissolved oxygen seemed to be unstable during the test period. Therefore, the BOD_5 tests were only available to phase 1 for more than 4 months, and then were terminated. Principles of such determinations are summarised in Table 3.3.

Table 3.3. Principle for determination of TOC, COD and fatty acids

Parameter	**Principle**	**Standard**
COD	Oxidisable substances react with sulphuric acid - potassium dichromate solution in the presence of silver sulphate as a catalyst. Chloride is masked by mercury sulphate. The green coloration of Cr^{3+} is evaluated.	ISO 6060-1989 or DIN 38409-H41-H44
BOD_5	The method consists of filling the samples to overflowing airtight bottles of the specified size and incubating them at the specified temperature (20 °C) for 5 days in the dark. Dissolved oxygen is measured initially and after incubation, and the BOD_5 is computed from the difference between initial and final dissolved oxygen values.	DIN EN 1899-1-H51
TOC	In a two-stage process, the total inorganic carbon (TIC) is first expelled with the help of the TOC - X5 shaker, then the TOC is oxidised to CO_2. The CO_2 passes through a membrane into the indicator cuvette, where it causes a colour change occur, which is evaluated with a photometer.	EN 1484 or DIN 38409-H3
Fatty acids	Fatty acids react with diols in an acidic environment, forming fatty acid esters. These esters are then reduced by iron (III) salts to form red coloured complexes, which are photometrically evaluated.	Hach Lange cuvette test kits LCK 365

3.2.2.4 Characteristics of the exhausted gas

Gas emission behaviours are used to evaluate the internal conditions of the lysimeters. They can be also the supplemental indicators in addition to the parameters of leachate for interpreting certain changes occurring inside the lysimeters.

Analyses of CH_4, CO_2, and O_2 were performed using the Gas Chromatography Shimadzu GC-8A.

3.2.3 Schematic of analytical procedures

For each phase of the experimental process, the sequence of preparatory and/or analytical procedures is similar and can be summarised in Figure 3.1 below. In general, the characterisation of each lysimeter is determined by investigating its solid, liquid (eluate and leachate), and gas phase. However, the main and long-term analytical investigations are focused on the concerned parameters of the leachate generated from each lysimeter. The unloading of test lysimeters were only available to phase 2.

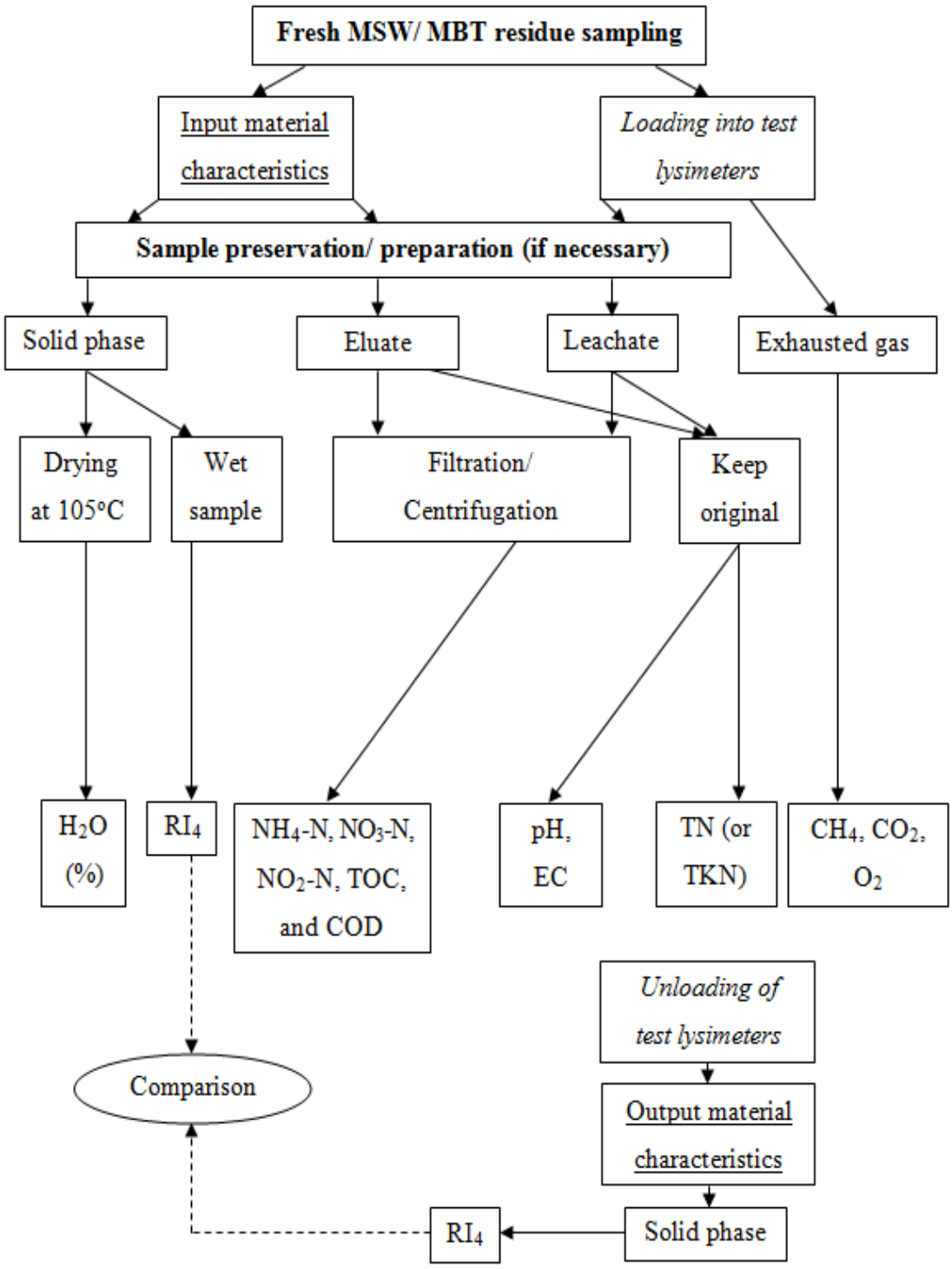


Figure 3.1. Schematic of preparatory and analytical procedures

3.3 Lysimeter settings and operations

In each lysimeter a leachate recirculation system has been installed, in which the leachate container is connected to the bottom of the lysimeter. Leachate is recirculated by mean of a peristaltic pump which timely runs thanks to a time switching device. The recirculated

leachate is distributed to the top of each lysimeter through three holes evenly located on the cap of the lysimeter. Gas collecting bags are connected to the lysimeters at the top covers. Regarding phase 2, gas collecting bags were not present and there was only one hole at the top of each lysimeter for the distribution of recirculated leachate due to the small diameter and volume of lysimeters being used. In the tests where aeration has been applied, aeration systems have been installed in both the solid waste and the leachate containers. The different boundary conditions will be mentioned in details later for each specific phase. Leachate samples were taken at the lysimeter's bottom in phases 1 and 2. In phase 3, leachate samples were directly collected through a valve joined with the leachate container.

3.3.1 Lysimeter settings for phase 1

In this phase, the fresh MSW was sampled at the MBT plant in Göttingen and loaded into the lysimeters on the same day. Whereas, the MBT residue was also sampled there but has been stored in two closed containers at room temperature for a couple of years. Characteristics of the fresh MSW and MBT residue for phase 1 are showed in Table 3.4.

Table 3.4. Characteristics of input materials for phase 1

Analysis	**Parameter**	**Unit**	**MBT residue**	**Fresh MSW**	**Ratio of MSW/MBT**
Solid phase	Water content	% on wet basis	27.7	50.9	1.84
	RI_4	mg O_2/g DM	5.8	49.7	8.57
Eluate (Leaching test)	NH_4-N	mg/L	18.5	36.4	1.97
	NO_3-N	mg/L	0.8	10.0	12.50
	NO_2-N	mg/L	< 0.6*	< 0. 6*	-
	TOC	mg/L	103	3,976	38.60
	COD	mg/L	347	10,947	31.55

*DM: dry matter; *: detection limit*

It can be clearly seen from Table 3.4 that MBT process has produced strong effects on the removal of organic carbon substances. The leaching tests showed that TOC and COD values in the eluate from fresh MSW were roughly 38.6 and 31.6 times higher than those in the eluate from MBT residue, whereas ammonium nitrogen concentration of the fresh MSW eluate is only nearly double that of the MBT residue eluate.

Four lysimeters were operated under similar conditions of room temperature and leachate recirculation. Their boundary conditions are shown in Table 3.5.

Table 3.5. Boundary conditions for four lysimeters in phase 1

Parameter		**Unit**	**MBT LSR 1**	**MBT LSR 2**	**MSW LSR 3**	**MSW LSR 4**
Waste mass in wet scale		kg	85.90	85.90	72.25	70.84
Filling density		kg/dm^3	0.88	0.94	0.75	0.76
Initial water volume added		L	44.0	44.0	28.5	28.0
Start of irrigation and leachate recirculation		day after loading	14	14	8	8
Recirculation rate	until day 101[(a)]	L/min	0.15	0.15	0.19	0.19
	from day 105[(b)]		0.19	0.19	0.19	0.19
Time of each recirculation		min	30	30	30	30
Recirculation interval		min	240	240	240	240
Start of leachate analysis		day after loading	25	25	11	11
End of the test		day after loading	295	295	250	250

[(a)]: Since day 0 (loading) until day 101; *[(b)]: From day 105 to the end of the test*

It is necessary to notice that the silicon tubes for leachate recirculation of two MBT lysimeters 1 and 2 was broken 87 days after the start of irrigation and leachate recirculation. The new tubes with a little larger diameter were then replaced, leading to a higher recirculation rate (0.19 L/min versus the former value of 0.15 L/min).

The schematic settings of the 141-L lab-scale lysimeters for phase 1 are illustrated in Figure 3.2.

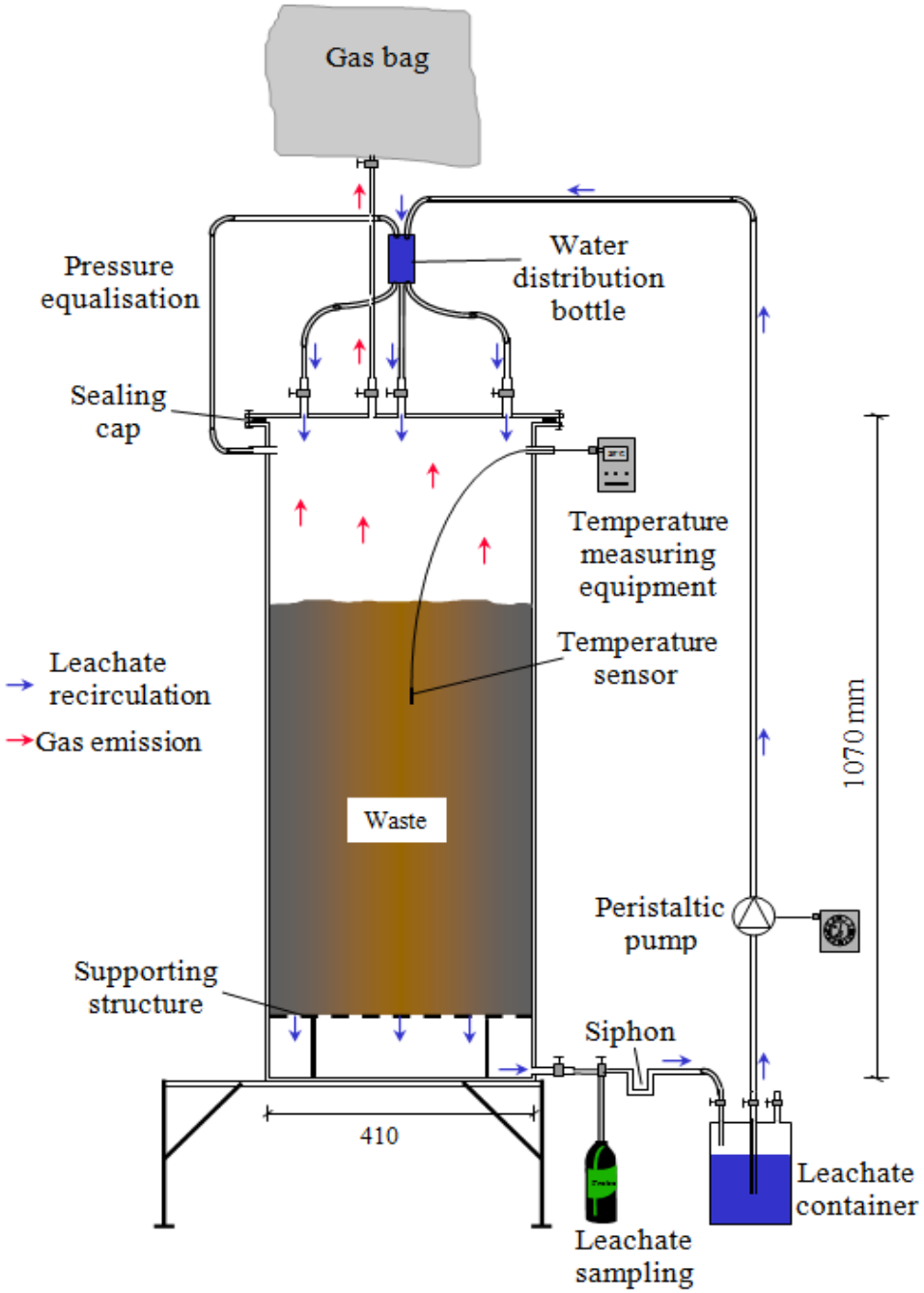


Figure 3.2. Schematic of the 141-L lab-scale lysimeter for phase 1

3.3.2 Lysimeter settings for phase 2

In this phase, the MBT residues have been also sampled at the MBT plant in Göttingen. They have been used as the input material for four small 1.34-L lysimeters. Characteristics of the material were determined by analysing the solid phase and its eluate (Table 3.6).

Table 3.6. Characteristics of input MBT residue in phase 2

Analysis	Parameters	Unit	Value
Solid phase	Water content	% by wet mass	27.3
	RI_4	mg O_2/g DM	5.9
Eluate (Leaching test)	NH_4-N	mg/L	21.8
	NO_3-N	mg/L	0.9
	NO_2-N	mg/L	<0.6*
	TOC	mg/L	121.0
	COD	mg/L	408.0

*: *detection limit*

All four small lysimeters (d = 10 cm; H = 17 cm) contained MBT residue. Two lysimeters 1 and 2 were operated with leachate recirculation (assigned as recirculation lysimeters). The lysimeters 3 and 4 were operated under the combination of intermittent aeration and leachate recirculation (assigned as hybrid lysimeters). Each hybrid lysimeter was intermittently aerated by a small aquarium pump. The intermittent aeration was simultaneously applied to both liquid and solid phases. The aeration for liquid is obviously easier than for solid because of the fact that solid waste always has a higher resistance to aeration. The schematic settings of these lysimeters are illustrated in Figure 3.3 below.

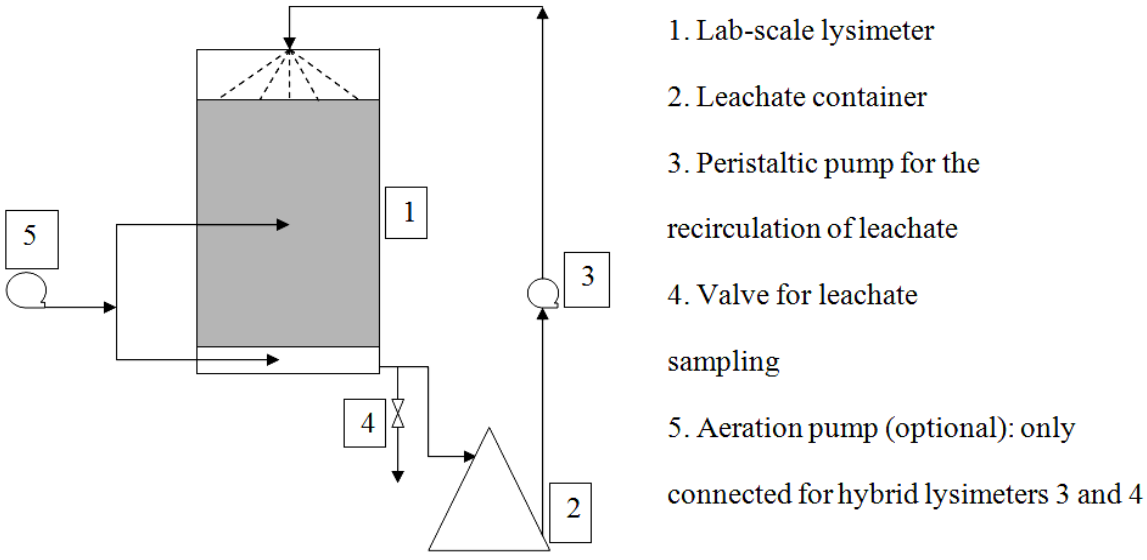


Figure 3.3. Schematic of 1.34-L small lab-scale lysimeters for phase 2

The boundary operational conditions of the small lysimeters are summarised in Table 3.7.

Table 3.7. Boundary operational conditions for the small lysimeters in phase 2

Parameters (unit)	**Recirculation lysimeters**		**Hybrid lysimeters**	
	LSR 1	**LSR 2**	**LSR 3**	**LSR 4**
MBT residue mass $_{\text{in wet scale}}$ (kg)	1.034	1.051	1.049	1.065
Loading density (kg/dm^3)	1.054	1.071	1.069	1.085
Time of each recirculation (min)	21	21	21	21
Recirculation interval (min)	180	180	180	180
Initial water volume added (L)	0.5	1.0	1.0	1.0
Daily recirculation leachate volume (L)	0.5	1.0	1.0	1.0
Initial Liquid/Solid $_{\text{wet}}$ ratio (-)	0.483	0.951	0.960	0.946
Start of aeration (LSR 3 and 4) after the residue being loaded for 7 days				
Aeration rate (L/min/kg waste)			0.192	0.246
Time of each aeration (min)/ Aeration interval (min)			20/ 720	20/ 360
Double aeration frequency after the start-up of aeration (LSR 3 and 4) for 14 days				
Aeration interval (min)			360	180
Total aeration time (day)			52	52

3.3.3 Lysimeter settings for phase 3

Phase 3 involves the settings and operation of six 141-L lysimeters. Two lysimeters (LSR 1 and 2) were loaded with MBT residue and four others (LSR 3, 4, 5, and 6) with MSW material. However, lysimeters 3 and 4 had a connection to phase 1. It is necessary to address their operation history. After the stop of four lysimeters in phase 1, two containing MBT residue (LSR 1 and 2) were unloaded, whereas two lysimeters (LSR 3

and 4) containing fresh MSW were kept closed and placed at the original place for approximately 7 months. They have been then moved to another place in the laboratory and continuously used for phase 3 of the research programme. Of which, lysimeter 3 has been run under leachate recirculation combined with aeration. In the meantime, lysimeter 4 has been continuously controlled in the manner similar to its previous conditions (e.g. by only leachate recirculation).

In lysimeter 4 the waste was not changed, whereas lysimeter 3 was opened to install an aeration system. In the lysimeters 1, 2, 5, and 6 aeration tubes were also installed.

The MBT residue loaded into lysimeters 1 and 2 is similar to that used for phase 2 at some extent. The fresh MSW loaded into lysimeters 5 and 6 is very similar to that loaded into lysimeters 3 and 4 in phase 1. Characteristics of the MBT residue and the fresh MSW are presented in Table 3.8. The old MSW in lysimeters 3 and 4 was not analysed to avoid the change in its mass and the internal anaerobic conditions.

Table 3.8. Characteristics of input MBT residue and fresh MSW for phase 3

Analysis	**Parameter**	**Unit**	**MBT residue**	**Fresh MSW**
Solid phase	Water content	% by wet mass	29.8	49.67
	RI_4	mg O_2/g DM	5.21	44.71
Eluate (Leaching test)	NH_4-N	mg/L	26.1	45.6
	NO_3-N	mg/L	0.98	12.1
	NO_2-N	mg/L	< 0.6*	< 0.6*
	TN	mg/L	47.0	80.4
	TOC	mg/L	115.7	4,266
	COD	mg/L	393.2	11,564

**: detection limit*

The fresh MSW material loaded into lysimeters 5 and 6 has been also sampled from the MBT plant in Göttingen, Germany.

The schematic view of lab-scale lysimeters of phase 3 is shown in Figure 3.4. It is in general similar to that of phase 2, but at much larger scale.

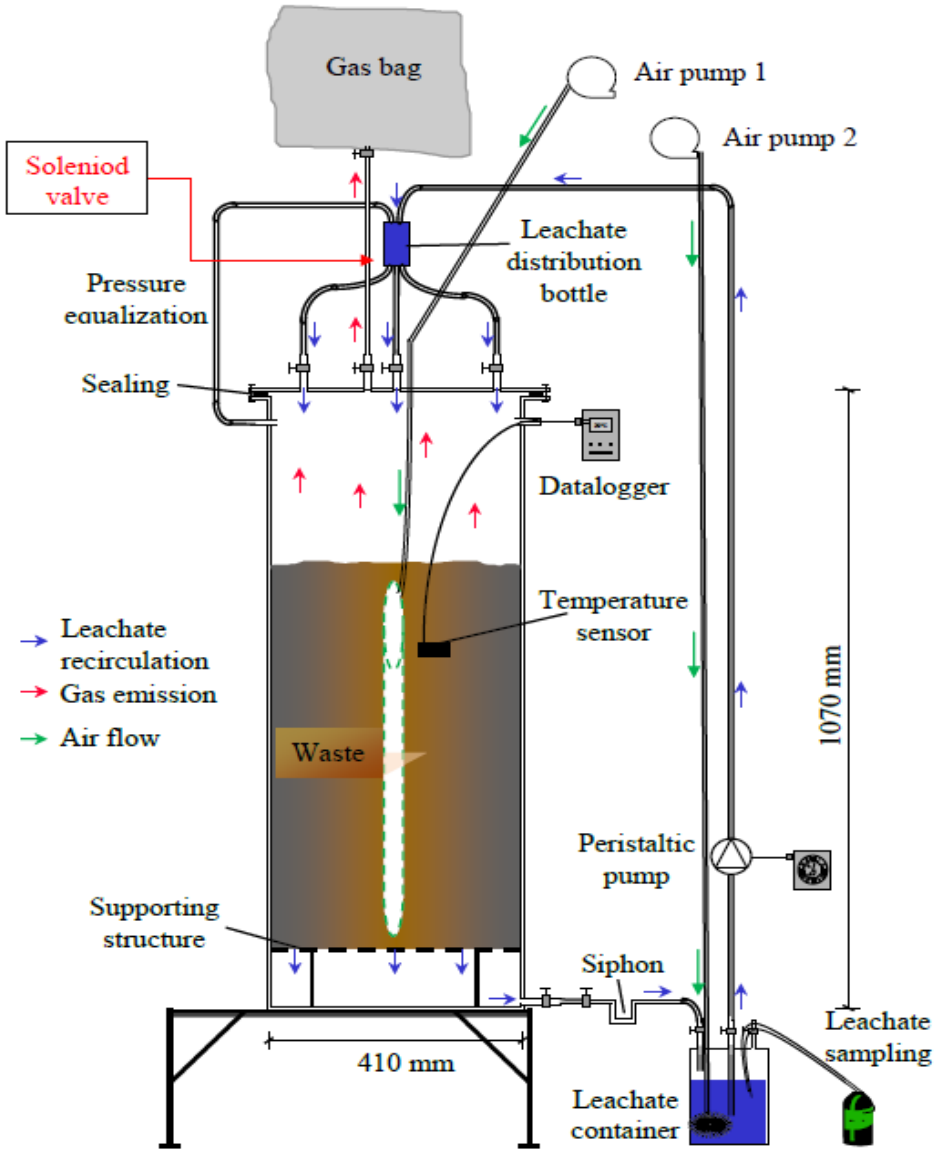


Figure 3.4. Schematic of the 141-L lab-scale lysimeters for phase 3 (LSR 4 has no aeration system and solenoid valve)

The real settings for phase 3 are similar to those for phase 1 in respect of leachate recirculation system; however, five lysimeters 1, 2, 3, 5, and 6 have been additionally installed aeration systems. Four lysimeters 1, 2, 3, and 4 were run under room temperature of about 20 °C, whereas two remainders (LSR 5 and 6) were operated in a warm chamber of about 30 °C. As aeration has been initiated, both aeration and recirculation regimes for LSR 6 have been intentionally set to occur at the same time. Meanwhile, such regimes for LSR 5 were taken place at different time. The solenoid valves were timely set to open for 15 minutes right before the lysimeters were aerated.

The purpose of this procedure is to limit the injected air entering into the gas bag. The boundary operational conditions are summarised in Tables 3.9 and 3.10.

Table 3.9. Boundary conditions for six lysimeters in phase 3

Parameter	**Unit**	**MBT LSR 1**	**MBT LSR 2**	**Old MSW LSR 3**	**Old MSW LSR 4**	**Fresh MSW LSR 5**	**Fresh MSW LSR 6**
Waste mass in wet scale	kg	81.30	85.17	72.09	70.84	65.62	59.60
Loading density	kg/dm^3	0.800	0.827	-	-	0.700	0.674
Loading & installing aeration system	day	0	0	0[(a)]	-	0	0
Temperature	°C	20	20	20	20	30	30
Start of recirculation	day after loading	11	11	11	11	5	5
Recirculation rate	L/60 min	7.17	6.93	6.93	6.13	13.6	13.6
Time of each recirculation	min	30	30	30	30	30	30
Recirculation interval	min	240	240	240	240	360	360
Initial water volume added to LSR 1, 2, 5 & 6 / *Leachate volume (LSR 3 & 4)*	L	29.1	29.4	*28.5*[(b)]	*28.5*[(b)]	27.25	24.75
Daily recirculation leachate volume	L	21.51	20.76	20.76	18.39	27.24	27.24
Test duration	day after loading	413	413	413	413	310	310

[(a)]: LSR 3 was installed aeration tube only; *[(b)]: Leachate volume (for LSR 3 and 4) was kept from phase 1.*

Table 3.10. Aeration history and parameters for five aerated lysimeters in phase 3

Aeration parameter/ Remark		**Unit**	**MBT LSR1**	**MBT LSR2**	**Old MSW LSR3**	**Fresh MSW LSR 5**	**Fresh MSW LSR 6**
Loading & installing aeration system		day	0	0	0[(a)]	0	0
Start aerating LSR 2 and 3 intermittently[(b)]		day after loading	-	27	27	-	-
Start aerating LSR 1 continuously		day after loading	96	-	-	-	-
Stop of aeration for LSR 2		day after loading	-	111	-	-	-
Stop of aeration for LSR 1/ Shift LSR 3 to continuous aeration		day after loading	133	-	133	-	-
Start aerating LSR 5 & 6 intermittently	for solid phase	day after loading	-	-	-	117	117
	for liquid phase	day after loading	-	-	-	122	122
Time of each aeration		min	20	20	20	30	30
Aeration rate	for solid phase ($x10^{-3}$)	L/min/kg waste	52.60	37.57	69.86	30.5	33.6
	for liquid phase	L/min	2.74	2.56	3.66	3.40	3.03
Aeration interval		min	180	180	180	360	360
Increase in aeration rate for the liquid phase of LSR 3 after 168 days of operation							
Aeration rate		L/min	-	-	6.22	-	-
Shift LSR 3 to intermittent aeration after 265 days of operation							
Increase in aeration rate for the solid phase of LSR 5 and 6 after 200 days of operation							
Aeration rate ($x10^{-3}$)		L/min/kg waste	-	-	-	182.9	201.3
Total aeration time		day	37	84	386	193	193

[(a)]: LSR 3 was installed aeration tube only; [(b)]: For both solid and liquid phase together

The changes in aeration regime for five aerated lysimeters are listed in Table 3.11.

Table 3.11. Changing events in operational conditions for five aerated lysimeters (LSR 1, 2, 3, 5, and 6) in phase 3

Time (day)	**MBT LSR 1**	**MBT LSR 2**	**Old MSW LSR 3**	**Fresh MSW LSR 5**	**Fresh MSW LSR 6**
0	Loading of MBT residue		Installing aeration system	Loading of fresh MSW	
5				Start of irrigation & recirculation	
11	Start of irrigation & recirculation	Start of irrigation & recirculation	Start of recirculation		
27		Start of intermittent aeration			
96	Start of continuous aeration	Keep aerating intermittently	Keep aerating intermittently		
111	Keep aerating continuously	Stop of the intermittent aeration			
117				Start aerating solid phase intermittently	
122				Start aerating liquid phase intermittently	
133	Stop of aeration		Shift to continuous aeration		
168			Increase the aeration rate for the leachate		
200				Increase the aeration rate for solid phase	
265			Shift back to intermittent aeration		
310				End of the test	
413	End of the test				

It is necessary to keep in mind that as a lysimeter starts to be irrigated and/or recirculated; it means that the leachate recirculation is kept throughout the progress of operation. The application of aeration regime (either intermittent or continuous) is purposeful to observe the behaviours of the lysimeters under the defined boundary conditions. Whenever the terms “keep aerating intermittently” or “keep aerating continuously” appear, it is understood that the aeration regime is combined with leachate recirculation.

3.4 Calculations

It is well known that concentrations of N- and C- compounds as well as other pollutants in the leachates from real landfills are typically expressed in mass per volume unit.

As investigating the leachate from a real landfill, the amount of leachate to be sampled is negligible compared with the whole leachate amount of the landfill. However, it is necessary to take into account this ratio for this issue as doing laboratory tests, especially at small scales.

In case of lab-scale lysimeters, the boundary conditions are different from those of real landfills. A certain amount (volume) of water has to be added to the lysimeters at the beginning of their operation in order to obtain the leachate for recirculation. If the lab-scale lysimeters have small volume of, for example, one or few litres, then the amount of leachate to be sampled (if few hundreds mL) should possesses a relatively significant proportion of the whole leachate volume. Additionally, the loss of leachate on every sampling event and the addition of water to keep liquid phase constant should also cause significant flushing and dilution effects. Ignoring such effect will definitely bring to an inexact reflection of the real leachate concentration. Therefore, the calculations for the conversion of mg/L into mg/kg DM unit are given here to avoid the dilution effect. The change in liquid/solid ratio over the experimental time was also integrated into the calculation.

The calculations are based on the following assumptions:

- The total mass of solid waste to be loaded and leachate generated remains unchanged. This total mass in fact slightly decreases due to the emission of gases and the volatilisation of the liquid phase. However, the gas emissions as well as volatilisations should have little mass in comparison to that of the solid phase.
- The solid phase has equal water content in the whole bulk mass of the waste;
- The mass of solid particles in each leachate sample is negligible;
- The density of the liquid included in the wet solid waste is always equal to that of leachate produced and the water added (and equal to 1 g/cm^3).

The solid waste to be loaded into the lysimeter has a wet mass of m_o (kg) and a water content of W_{H2O} (%). The total volume of liquid phase is V_o (L) (= the sum of volume of water added at the beginning of the test plus the volume of liquid included in the wet solid waste which is $m_o*W_{H2O}/100$).

Table 3.12 expresses the procedures to convert the original concentration unit of mg/L into mg/kg DM.

Table 3.12. Interpretation of the calculation procedures

Sampling event	$\mathbf{V_{leachate\ out}}$	$\mathbf{V_{water\ in}}$	**Dilution factor**	$\mathbf{C_{measured}}$ **(mg/L)**	$\mathbf{C_{calculated}}$ **(mg/L)**	$\mathbf{C_{calculated}}$ **(mg/kg DM)**
1	$V_{1\ out}$	$V_{1\ in}$	$f_1 = 1$	$C_{v1.mes}$	$C_{v1.cal}$	$C_{m1.cal}$
2	$V_{2\ out}$	$V_{2\ in}$	$f_{2\ =}\ V_o/(V_o\text{-}V_1)$	$C_{v2.mes}$	$C_{v2.cal}$	$C_{m2.cal}$
...	.....	.....	.....	.....	.....	.....
i-1	$V_{i\text{-}1\ out}$	$V_{i\text{-}1\ in}$	$f_{i\text{-}1}= V_o/(V_o\text{-}V_{i\text{-}2})$	$C_{vi\text{-}1.mes}$	$C_{vi\text{-}1.cal}$	$C_{mi\text{-}1.cal}$
i	$V_{i\ out}$	$V_{i\ in}$	$f_i = V_o/(V_o\text{-}V_{i\text{-}1})$	$C_{vi.mes}$	$C_{vi.cal}$	$C_{mi.cal}$

Note: $V_{leachate\ out}$: volume of leachate being sampled; $V_{water\ in}$: volume of water being compensated; f: dilution factor; $C_{v.mes}$: concentration being measured; $C_{v.cal}$: concentration being calculated to avoid dilution effects; $C_{m.cal}$: concentration being calculated to avoid dilution effects.

$C_{v1.mes} = C_{v1.cal}$ is the concentration as the first specimen is sampled. $V_{1\ out}$, $V_{2\ out}$,..$V_{i\ out}$ are volume (L) of each specimen sampled on the *first*, *second*, ..., and *i* sampling event, respectively. Similarly, $V_{1\ in}$, $V_{2\ in}$, $V_{i\ in}$ are volume (L) of water added to the liquid phase of the lysimeter after each mentioned sampling event, accordingly (to keep the liquid phase constant). It means that $V_{1\ out} = V_{1\ in}$, $V_{2\ out} = V_{2\ in}$, ..., and $V_{i\ out} = V_{i\ in}$.

The mass of dried matter (m_{DM}) will be calculated as follows:

$$m_{DM} = \frac{m_o(100-W_{H2O})}{100}$$

Due to the loss of leachate on every sampling event and the addition of an equivalent water volume to the leachate container (dilution effect), the real concentration (in mg/L)

of a component in the leachate will be calculated based on its measured concentration (in mg/L) as follows:

$$C_{v1.cal} = C_{v1.mes} * f_1$$

$$C_{v2.cal} = C_{v2.mes} * f_2$$

.....

$$C_{vi.cal} = C_{vi.mes} * f_i$$

And by converting the unit from mg/L to mg/kg DM, the concentration of a component in the leachate for the sampling event 1, 2, .., and i ($C_{m1.cal}$, $C_{m2.cal}$...and $C_{mi.cal}$) are calculated as follows:

$$C_{m1.cal} = \frac{C_{v1.cal} * V_o * 100}{m_o * (100 - W_{H2O})} = \frac{C_{v1.mes} * f_1 * V_o * 100}{m_o * (100 - W_{H2O})} \text{ [mg/kg DM]}$$

$$C_{m2.cal} = \frac{C_{v2.cal} * V_o * 100}{m_o * (100 - W_{H2O})} = \frac{C_{v2.mes} * f_2 * V_o * 100}{m_o * (100 - W_{H2O})} \text{ [mg/kg DM]}$$

.....

$$C_{mi.cal} = \frac{C_{vi.cal} * V_o * 100}{m_o * (100 - W_{H2O})} = \frac{C_{vi.mes} * f_i * V_o * 100}{m_o * (100 - W_{H2O})} \text{ [mg/kg DM]}$$

From now on, the concentration of a component in the leachate generated from experimental LSR mentioned in the following chapters will be understood as $C_{mi.cal}$ (expressed in mg/kg DM).

CHAPTER 4: INFLUENCE OF LEACHATE RECIRCULATION ON THE QUALITY OF LEACHATES GENERATED FROM FRESH MSW AND MBT RESIDUE

4.1 Introduction

It has been stated in the literature by many authors that recirculation of leachate could enhance the stabilisation of old MSW, the gas production rate and leachate quality in MSW landfills (Pohland, 1980; Tittlebaum, 1982; Kinman et al., 1987; Doedens and Cord-Landwehr, 1989; Otieno, 1994; Townsend et al., 1996; Chugh et al., 1998; Onay and Pohland, 1998; El-Fadel, 1999; Pohland and Kim, 1999; San and Onay, 2001). The question is whether such technique could be applicable to the fresh MSW which typically have very high levels of several pollutants. To evaluate the application capability of this technique to the real conditions of fresh MSW landfills, laboratory tests were established to investigate the effect of leachate recirculation on the quality of leachate generated from fresh MSW. A similar procedure was also performed on MBT residue to assess whether such technique could bring about any improvement to the MBT residue which is already well stabilised after MBT process.

In this chapter, the analytical results of the leachates generated from two fresh MSW lysimeters and two MBT lysimeters are presented. The results are used to evaluate the influence of leachate recirculation on the leachate characteristics.

As mentioned in Chapter 3, it must be kept in mind that the time for starting irrigation and recirculation as well as for analysing the concerned parameters regarding two fresh MSW lysimeters 3 and 4 are different from those as for two MBT lysimeters 1 and 2.

4.2 pH and electrical conductivity values

4.2.1 pH value

The experimental results clearly show that pH values for the leachate from two fresh MSW lysimeters increase from about 5.0 to 6.5 during the first 4 weeks of operation (Figure 4.1). The increase in pH in this duration should be attributed to the acclimatisation of microorganisms, by the pH neutralisation of the added water (i.e. dilution effect), and the recirculation of leachate.

As the MSW inside lysimeter become more saturated, the microbes grow slowly, which lead to no more raising pH levels. Additionally, due to the hydrolysis of the

biodegradable fraction of the solid waste (and other applied liquids), volatile fatty acids (VFA) are accumulated in the leachate, which results in a decrease in pH.

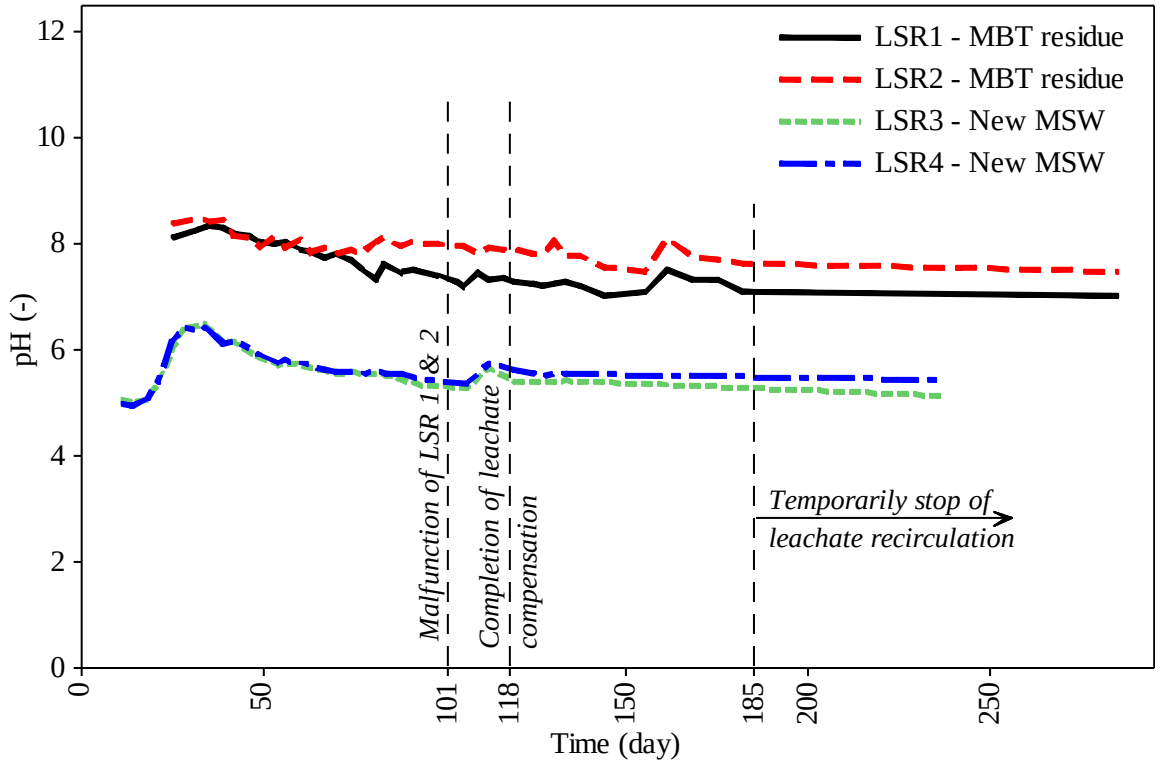


Figure 4.1. Variations in pH values of the leachate

The experimental operation observed that both pH curves for fresh MSW lysimeters 3 and 4 seem to coincide, proving that the very similar environmental conditions were obtained inside the waste mass of the two lysimeters. The long-term average pH values for these two lysimeters are of approximately 5.5. These values are in the pH range of landfill leachate in acid forming phase (4.5 – 7.5). These pH values are unfavourable for the anaerobic microorganisms to grow as the optimal pH range for their growth is from 6.0 to 8.0 (Ehrig, 1983).

The pH values for the MBT lysimeters are much higher, ranging from around 7.0 to 8.3 and 7.5 to 8.5 regarding lysimeters 1 and 2, respectively. In comparison to literature, these values relatively agree to the pH value of landfill leachate in methanogenic phase (7.5 – 9.0). There was a slight increase in pH values for both MBT lysimeters in the first month of operation. This increase can be explained similarly to what happened to two fresh MSW lysimeters. However, the MBT residue has much less organic matters in its composition compared with the fresh MSW, leading to the less increasing pH intensity and higher pH values.

4.2.2 Electrical conductivity

The measurements (Figure 4.2) show that electrical conductivity (EC) values of the leachates from all lysimeters are much higher comparing with the data from the literature (see Table 2.1 in section 2.3.2.2). EC values in the leachates generated from two MBT lysimeters 1 and 2 were stabilised at about 30 mS/cm from the beginning of the operation until day 101 when the significant dilution event occurred.

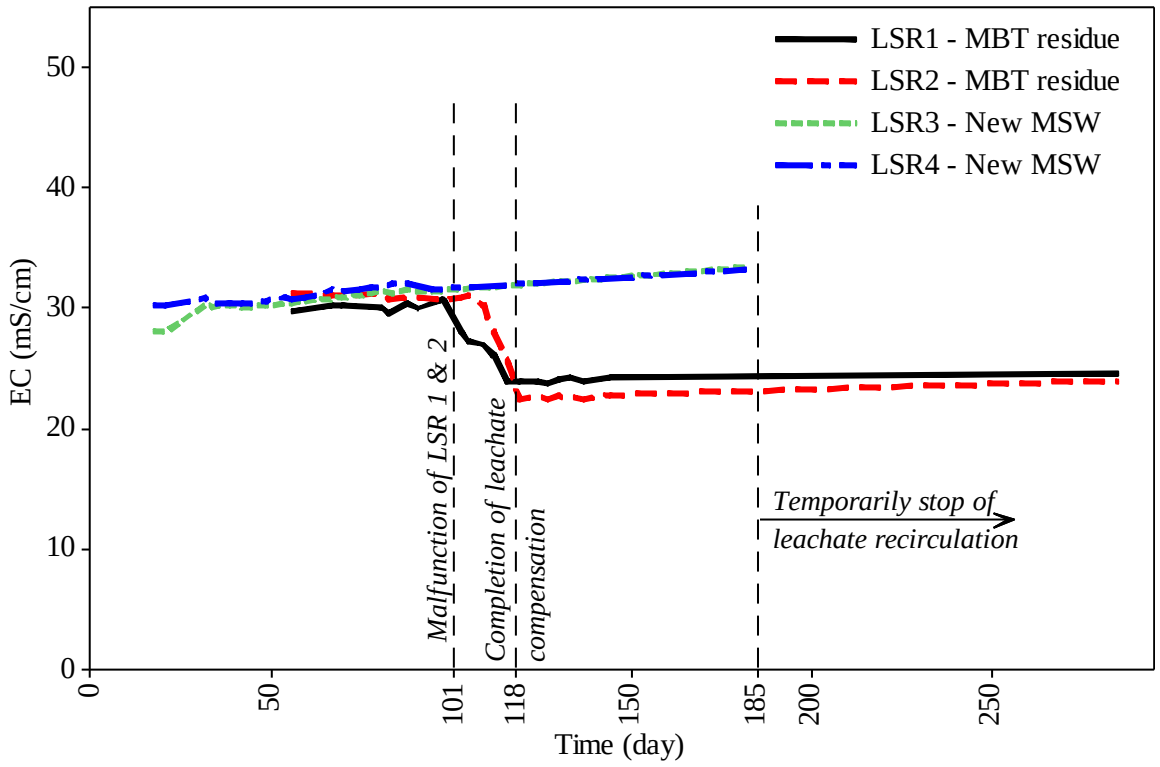


Figure 4.2. Variations in EC values of the leachate

Due to an unexpectant malfunction during the experimental run (day 101 after loading), the silicon recirculation tubes of two MBT lysimeters 1 and 2 were broken, leading to the loss of leachate volumes of 8 and 17.5 L, respectively. Equivalent water volumes were then accordingly compensated to the leachate containers. The higher flushing and dilution effects in case of lysimeter 2 evidently resulted in its constantly lower EC values comparing with those of lysimeter 1 after the completion of leachate compensation.

The EC values of two fresh MSW lysimeters 3 and 4 were slightly increased from the start-up of analyses and stably fluctuated in the range from 31 to 32 mS/cm. It can be assumed that the EC value differences between two fresh MSW lysimeters and two MBT residue ones would be low if the breakdown did not happen to two MBT lysimeters.

4.3 Nitrogen parameters

4.3.1 Ammoniacal nitrogen

The experimental results of this study clearly show the huge differences regarding the concentrations of ammonium nitrogen in the leachate from MBT residue and fresh MSW (Figure 4.3).

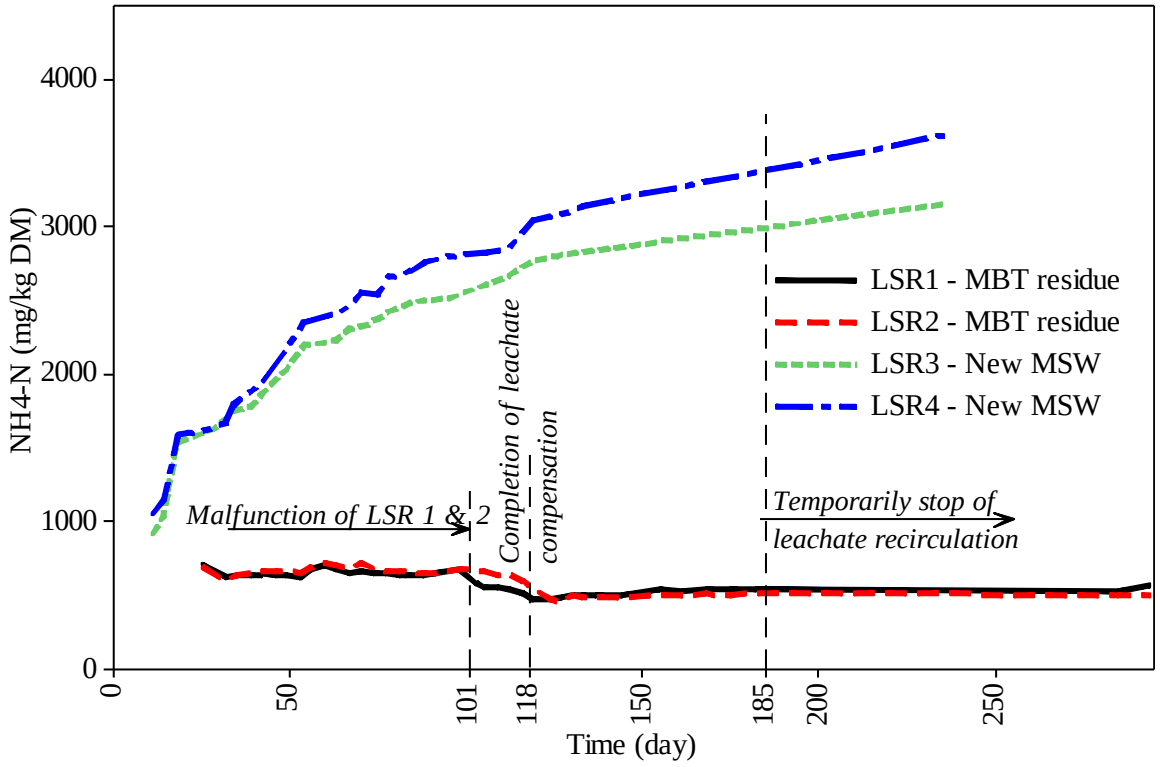


Figure 4.3. Variations in concentration of ammonium nitrogen in the leachate

Ammonium nitrogen concentration in the leachate from MBT residue is stably fluctuating at about 660 mg/kg DM during first 14 weeks after the start-up of their operation. There was then a relatively significant decrease in its concentration (after day 101). The reason is that the silicone recirculation tubes for both lysimeters 1 and 2 during this time were broken, leading to the losses of leachate. Equivalent volumes of tap water then have been filled back to each leachate container. This dilution event could be visibly seen through the sharply abnormal decline of EC values (Figure 4.2 in section 4.2.2).

Additionally, the gas collection bag of lysimeter 1 was broken, also leading to either a loss of gases or a penetration of the air inside the lysimeter. The malfunction might affect the concentration of ammonium nitrogen due to nitrification, which will be discussed later in more details on section 4.3.2. For a better illustration of these effects only the

concentrations of ammonium nitrogen in the leachates from the MBT lysimeters 1 and 2 are shown in Figure 4.4.

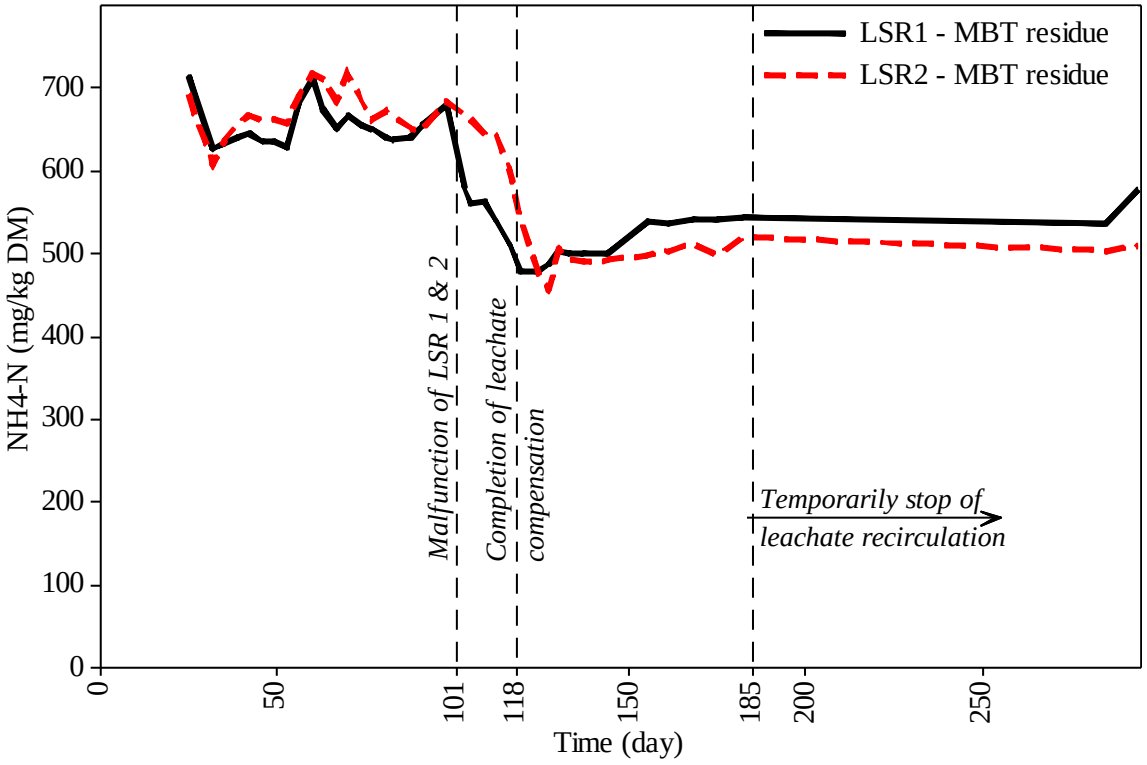


Figure 4.4. Variations in concentration of ammonium nitrogen in the leachates generated from LSR 1 and 2

It can be clearly seen from Figure 4.4 that ammonium nitrogen concentration in the leachate from lysimeter 1 after the completion of leachate compensation was almost higher than that of lysimeter 2 with higher flushing and dilution effect.

Basically, the two curves for ammonium nitrogen concentrations in the leachates from lysimeters 1 and 2 are nearly identical. There were sometimes slight increases in ammonium concentrations, which partially imply that very low (or extremely low) ammonification process occurred inside the two MBT lysimeters.

As for two fresh MSW lysimeters, ammonium nitrogen concentrations in their leachates dramatically increased after the start-up of experimental operation. The starting values were at about 915 and 1,048 mg/kg DM for the leachate generated from lysimeters 3 and 4, respectively. These values rapidly rose up to approximately 1,500 mg/kg DM in the first week. The fast increase in ammonium concentration addresses that the recirculation of leachate has steadily promoted the ammonification as well as flushing and/or leaching process. The two concentration curves had a similar tendency. However, it is observed that ammonium concentration in the leachate from lysimeter 4 is almost higher than that

from lysimeter 3. This indicates that the fresh MSW material is still heterogeneous at some extent, despite its previous shredding for size reduction.

4.3.2 Nitrite and nitrate nitrogen

During the entire tests, concentrations of nitrite nitrogen in the leachates from three lysimeters 2, 3, and 4 were under the detection limit of the analysis (< 0.6 mg/L N-NO_2 by using Hach Lange cuvette test kits LCK 342). It is comprehensible since the nitrification process should almost have not occurred because of the fact that these three lysimeters were operated under anaerobic conditions. However, lysimeter 1 showed some variations in nitrite nitrogen concentration (12.3 and 14.1 mg/kg DM on day 105 and 109, respectively). That can be attributed to the fact that the lysimeter had the air-ingress problem as it was partially malfunctioning.

The variations in concentration of nitrite nitrogen in the leachate from lysimeter 1 and of nitrate nitrogen from all four lysimeters are shown in Figure 4.5.

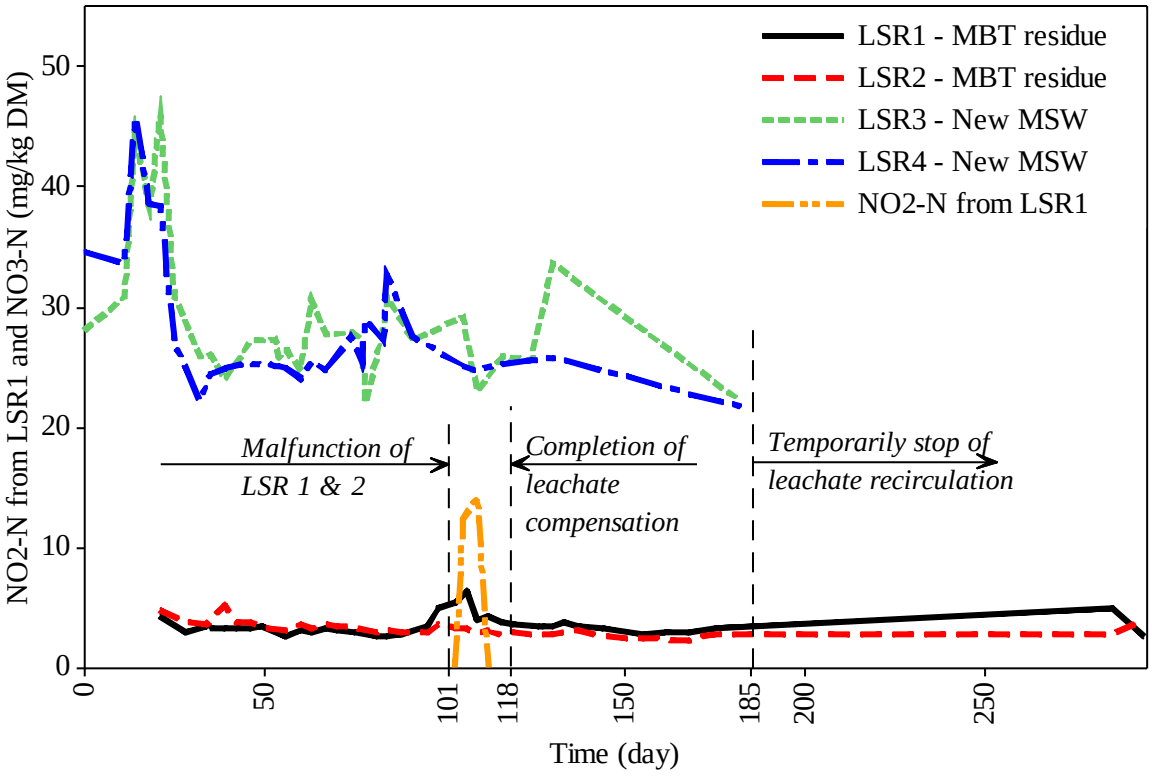


Figure 4.5. Variations in concentration of nitrate in the leachate from four lysimeters and nitrite nitrogen in the leachate from LSR 1

It can be seen that the concentrations of nitrate nitrogen in the leachate from lysimeter 2 were comparatively stabilised at about 3.5 mg/kg DM. Such stable nitrate nitrogen levels in combination with the data of ammonium nitrogen in its leachate suggested that nearly

no nitrogen conversions had occurred in this lysimeter. In case of lysimeter 1, the malfunction event on day 101 has led to the change from anaerobic to aerobic at some extent. A short and a little rise in nitrate and nitrite nitrogen concentrations was an evidence of such change. The changes in nitrite and nitrate nitrogen values as well as the steady downhill course of ammonium nitrogen concentrations after the malfunction event (see Figures 4.4 and 4.5) strongly confirm that nitrification had shortly taken place.

A fast and short increase in nitrate nitrogen concentrations in lysimeters 3 and 4 at the beginning of their operation might have also been due to the effect of nitrification. The process might have occurred due to the fact that there was a small certain amount of oxygen in the air during the filling of the waste material. However, the rate of nitrification and the number of nitrifying bacteria would have been limited because the pH values in this duration as well as in the whole experimental period were almost less than 6.0. Such nitrification process would have also happened to lysimeters 1 and 2 after their start-up for a couple of days, but in these tests leachate probes were sampled just after beginning of the operation.

The nitrate nitrogen concentration curves in lysimeters 3 and 4 are not smooth like those regarding lysimeters 1 and 2, but rather fluctuating, partly implying that the fresh MSW materials are relatively more heterogeneous.

A combination of the low and stable concentration of nitrate nitrogen and the low and stable concentration of ammonium nitrogen in the leachates from MBT lysimeters 1 and 2 (comparing with those from MSW lysimeters 3 and 4) could imply that a major proportion of organic nitrogen compounds in the solid MSW (the MSW input that originates the MBT residue loaded into two MBT lysimeters 1 and 2) have been converted to other nitrogen forms during MBT process. Many possible nitrogen conversions may take place during the biological treatment that is integrated into MBT, and most of these are yet not well-understood. Different processes, such as mineralisation, volatilisation of ammonia gas due to high temperatures achieved during composting processes, nitrification and denitrification with various routes, as well as incorporation into stable organic compounds can effectively change nitrogen content. However, it was stated in literature that high ammonia gas as well as nitrous oxide emissions from MBT plants during biological treatment have been observed (Fricke et al., 2005).

4.3.3 Total Kjeldahl nitrogen

The analyses showed TKN values which were almost slightly higher than those of ammonium nitrogen. There were, however, some points in which the TKN was less than

ammonium nitrogen concentration. This illogical fact has caused by the analytical error as digesting the leachate sample for determination of TKN. During the digestion of the sample at high temperature in the presence of concentrated sulphuric acid and hydrogen peroxide mixture, whenever the experimental manipulation control was not well performed, it would lead to the overflow and loss of the sample.

4.4 Organic carbon parameters

4.4.1 Chemical and biochemical oxygen demands

During the whole tests a huge gap between the values of COD in the leachates from fresh MSW and MBT lysimeters was observable (Figure 4.6). It is necessary to notice that the COD concentration showing in the figure is in logarithmic scale.

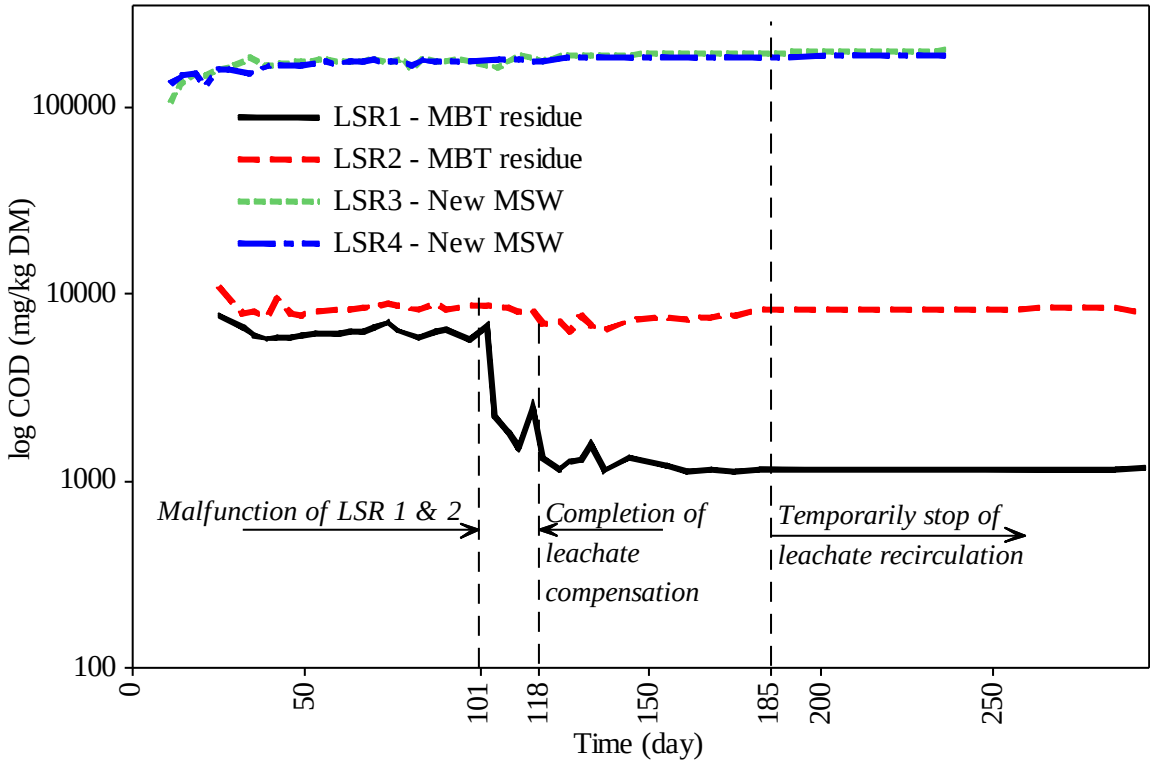


Figure 4.6. Variations in concentration of COD in leachate

The initial COD concentrations of the leachates from MBT lysimeters 1 and 2 were 7,595 and 10,818 mg/kg DM, respectively. Whereas, these values regarding MSW lysimeters 3 and 4 were more than 10 times higher at 105,313 and 131,945 mg/kg DM, respectively.

The sudden and dramatic decrease in COD in the leachate from lysimeter 1 can be again explained by the accidentally unexpected malfunction occurring to the lysimeter on day 101. Additionally, a strong malodour of hydrogen sulphur gas was qualitatively detected

by the sense of smell as opening the lysimeter 1's leachate container cap during this event. Such behaviours could likely suggest that sulphate or sulphite might be used by sulphate reducing bacteria as electron acceptors during the oxidation of organic materials under anaerobic condition to form the end product hydrogen sulphur gas regarding the lysimeter 1. The analysis of sulphate was not subject of the tests so that there is not enough persuadable evidence to be firmly sure of that assumption.

However, the analyses on the exhausted gas composition from lysimeter 1 during this period also showed certain differences from the others. Carbon dioxide was slightly risen up; meanwhile, methane was increased to 21 % on day 144 and reached the value of about 44.3 % on day 161. Such behaviours should have confirmed the onset of methanogenic condition in the lysimeter which contributed to the reduction of COD at certain degree.

Lysimeter 2 has also experienced a breakdown problem leading to the loss of leachate, but the COD values showed a totally unlike tendency. The specific stink of hydrogen sulphur gas was not smelt in the leachate container. Nevertheless, no smelling of hydrogen sulphur does not mean it is not emitted because hydrogen sulphur has no perceivable odour at dangerously high concentrations. Moreover, the analyses of the exhausted gas composition from lysimeter 2 showed no methane, addressing that the methanogenic condition was not created in the lysimeter.

As for lysimeters 3 and 4 containing fresh MSW, the concentrations of COD significantly increased and reached a quite stable level after about 50 days since the beginning of their operation.

As for BOD_5 parameter, the calculated values basing on experimental data show highly fluctuated values (Figure 4.7).

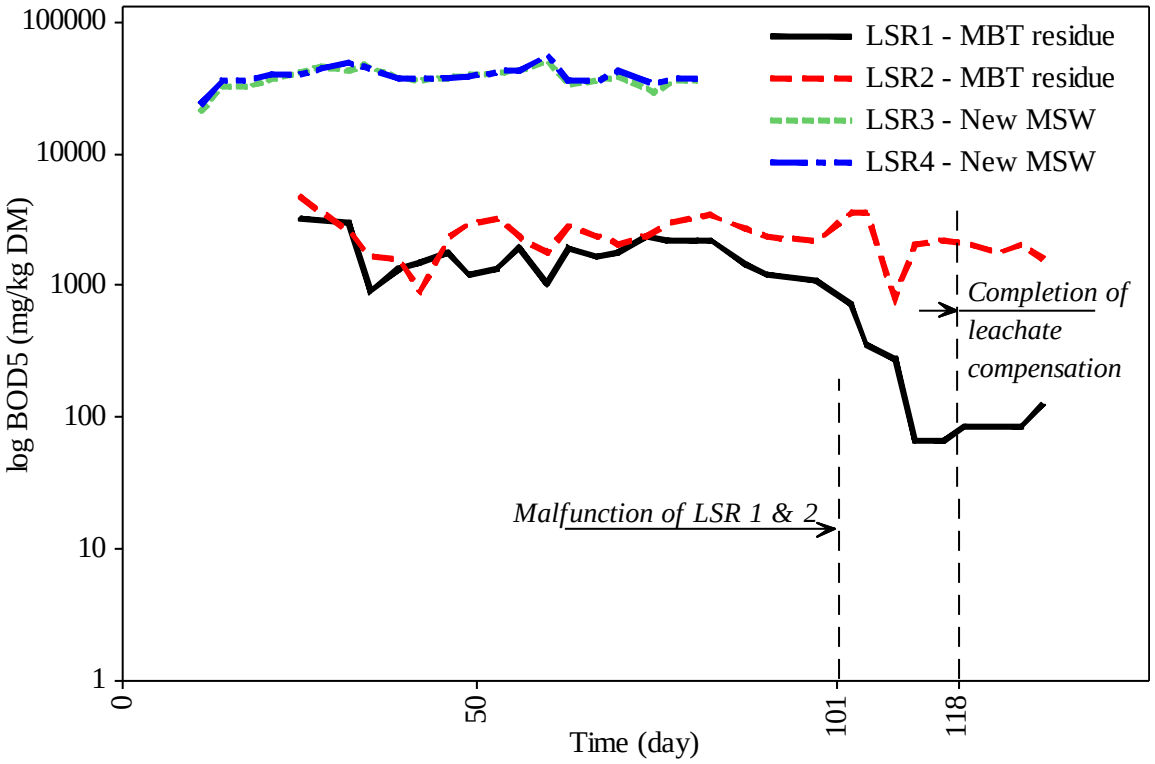


Figure 4.7. Variations in concentration of BOD_5 in leachate

The minimum BOD_5 values for the leachates from lysimeters 1, 2, 3, and 4 are 67, 804, 21,050, and 24,050 mg/kg DM, respectively, whereas the maximum values are 3,310, 4,810, 51,040, and 57,040 mg/kg DM, respectively. Due to the unstable behaviour of the dissolved oxygen sensor (already mentioned in section 3.2.2.3), the highly varying BOD_5 values obtained as well as the consumption of time regarding the determination procedure, the BOD_5 tests have been terminated after 130 days of operation.

4.4.2 Total organic carbon

TOC is an indicator of leachate organic strength in addition to COD. It clearly shows again that there was an enormous gap between the values of TOC in the leachates from MBT residue and fresh MSW (Figure 4.8). The initial TOC concentrations of the leachates from MBT lysimeters 1 and 2 were 2,808 and 3,590 mg/kg DM, respectively; whereas those values regarding the leachates from fresh MSW lysimeters 3 and 4 were about 12 times higher at 33,730 and 43,122 mg/kg DM, respectively. TOC concentrations in the leachates from two fresh MSW lysimeters 3 and 4 were rapidly increased from their initial above-mentioned values to the levels of 52,656 and 50,493 mg/kg DM, respectively after 8 weeks since the start-up of their operation.

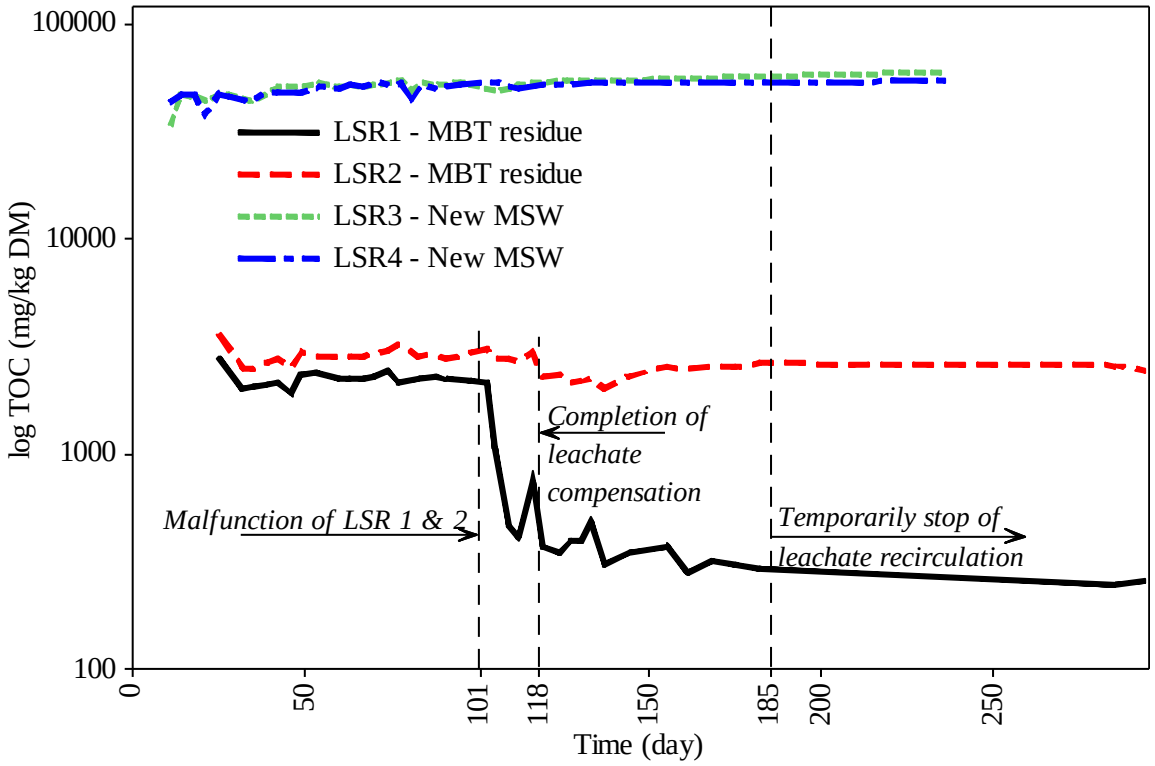


Figure 4.8. Variations in concentration of TOC in leachate

As for lysimeter 1, the average TOC value before the malfunction event on day 101 was about 2,300 mg/kg DM. It was then fast decreased to low stabilised level of about 370 mg/kg DM. The reasons for the swift decline in TOC values of the leachate from lysimeter 1 after the breakdown are explained in a similar way as the dramatical decrease in COD. The activities of methanogenic bacteria were firmly verified by the exhausted gas composition. Moreover, sulphate reducing bacteria were most likely existing in the system due to the detectable odour of hydrogen sulphur.

Lysimeter 2 had an average TOC of approximately 2,868 mg/kg DM before the breakdown. The TOC concentration was then slightly decreased. The lowest concentration of TOC for the leachate from lysimeter 2 was at 2,002 mg/kg DM on day 137 after the start-up of operation. It then slightly increased again and reached a value of 2,553 mg/kg DM on day 155. The concentration of TOC was, afterwards, quite stabilised at about 2,500 mg/kg DM. Although lysimeter 2 had a similar problem of leachate loss, the concentration of TOC in its leachate showed a different behaviour. The relatively stable data of TOC and COD in the leachate from lysimeter 2 and the gas composition without methane and nearly no carbon dioxide could address that methanogenic and sulphate reducing bacteria would have been almost inhibited.

4.4.3 COD/TOC ratio

As mentioned in Chapter 2 the COD/TOC ratio is indicative of the extent of oxidation of the organic compounds involved, in which a decrease in this ratio corresponds to a lower oxygen demand due to further oxidation of the organic matter.

Cameron and McDonald (1982) reported that the maximum possible COD/TOC is about 4.0 for relatively young fills, and it can be as low as 1.16 for old fills.

As for the test lysimeters, the experimental results generally agree well with the literature values. The COD/TOC ratios for the leachates from the two MSW lysimeters are stable around the value of 3.4, meanwhile this ratio fluctuates around 3.0 in case of two MBT lysimeters.

4.4.4 Variations of nitrogen and organic carbon parameters in the leachate from MBT residue and the biodegradability of the solid residue

The removal efficiency of ammonium nitrogen, COD, and TOC in the leachates produced from MBT residues is calculated based on the difference between values of such parameters measured at the first and the last sampling event (Table 4.1).

Table 4.1. Ammonium nitrogen, COD, and TOC in the leachate from MBT lysimeters

Parameter		**LSR 1**	**LSR 2**	**Removal efficiency (%)**	
				LSR 1	***LSR 2***
NH_4-N (mg/kg DM)	The first sampling	714.7	691.7	19.2	26.4
	The last sampling	577.3	508.9		
COD (mg/kg DM)	The first sampling	7,595	10,818	84.5	27.2
	The last sampling	1,176	7,879		
TOC (mg/kg DM)	The first sampling	2,808	3,590	90.9	31.7
	The last sampling	256	2,451		

The values from Table 4.1 show that the removal efficiency of ammonium nitrogen in the leachate from lysimeter 1 is much less than that of COD and TOC. Meanwhile, it is about the same range for such parameters in case of the leachate from lysimeter 2. Despite the

lysimeter 2 had higher dilution effects compared with the lysimeter 1 after the malfunction event (17.5-L versus 8-L of water compensation), the removal efficiency of both COD and TOC in the leachate from lysimeter 2 (27.2 and 31.7 %, respectively) is much less than that regarding lysimeter 1 (84.5 and 90.0 %, respectively). The air access into the lysimeter 1 also by the malfunction should have led to the conversion of organic carbon matters in the system, resulting in the high COD and TOC removal efficiency. Therefore, it is most likely that the effect of air ingress is far higher than the dilution effect.

The pair values of COD as well as TOC concentration in the leachates from two MBT residue lysimeters 1 and 2 show a similar developing trend prior to the malfunction event. The experimental results (depicted in Figures 4.5 and 4.7) show totally different tendencies after day 105 of their operation (4 days after the malfunction event).

In considering the biodegradability of MBT residues, RI_4 values regarding the loading and unloading residues for lysimeters 1 and 2 were calculated and shown in Table 4.2.

Table 4.2. Biodegradability of the MBT residue

Parameter	**Unit**	**Input value**	**Output value**	
			MBT LSR 1	***MBT LSR 2***
RI_4	mg O_2/g DM	5.76	2.58	3.89

It is calculated that the RI_4 values for unloading MBT residues from lysimeters 1 and 2 decreased by 55.2 and 32.5 %, respectively.

The reduction of RI_4 value should be due to flushing and/or leaching and dilution effects. However, the lower RI_4 output value in case of the lysimeter 1 once again indicates that there was a progressive reduction of biodegradable content in the system, which should be caused by the air access during its malfunction event.

4.5 Summary

The obtained results, in general, show slight effects of recirculation on the quality of leachates released from both types of the waste material during the experimental period.

With respect to two MBT lysimeters, all testing parameters seemed to be stabilised prior to their temporarily unexpected malfunction. The pH values of their leachates were very little affected by such short breakdown. It was observed the significant decreases in EC

values and in concentrations of ammonium nitrogen, TOC, and COD of their leachates after the abovementioned event. The values of these parameters, however, became fairly stable again as the leachate compensation was completed. Such incredible declines should have been mainly due to the unexpectant dilutions that have led to the strong flushing effect.

As for two fresh MSW lysimeters, the concentrations of ammonium nitrogen, TOC, and COD in their leachates have rapidly increased from the beginning to the end of their operation. The increased intensity in the concentration of these parameters was high at the start-up and to be moderate as their operation progressed. The regular dilutions (water compensation after each sampling) seemed to make no reduction effect on the concentrations of these parameters, as well as the EC values. This should be attributed to the fact that the flushing and/or leaching process was continuously taking place.

CHAPTER 5: NITROGEN AND ORGANIC CARBON REMOVAL FROM MBT RESIDUES BY LEACHATE RECIRCULATION IN COMBINATION WITH INTERMITTENT AERATION - A SMALL SCALE LYSIMETER STUDY

5.1 Introduction

From the literature relating to the stabilisation of MSW landfills, it is well known that recirculation of leachate and supplementary aeration can significantly reduce the landfill emissions in comparatively short time (Prantl et al., 2006; Ritskowski et al., 2006; Erses et al., 2008). While the effect of leachate recirculation is well known for MBT residue, the additional aeration has not been investigated. And so far, studies relating to the effects of such combination technique on MBT residue have not been officially published.

In a series of tests in laboratory scale the efficiency of this technique for MBT residue was investigated for different boundary conditions. Details on these conditions were already mentioned in section 3.3.2. The results show that this technique has only a limited influence on the reduction of organic compounds. In view of nitrogen compounds, only the additional aeration during recirculation shows a strong effect on the quality of leachate, in which the concentrations of ammonia and total nitrogen have been reduced by up to 99 %. The results indicate that by simple techniques the long-term emission behaviour of deposited MBT residue can be reduced fast to an acceptable level.

5.2 Nitrogen parameters

5.2.1 Ammonium

Under anaerobic conditions inside the landfill body, stabilisation process is relatively slow (Hudgins and March 1998) and there is no pathway for the transformation of ammonium (Berge et al. 2006). This results in the excessively slow decline of ammonium in the leachate over a long period of time.

The experimental results with MBT residues showed that ammonium nitrogen concentrations in the leachates from two recirculation lysimeters were relatively stable at around 600 mg/kg DM (Figure 5.1). A slight decrease in ammonium nitrogen concentrations under anaerobic conditions could be attributed to the dilution and flushing effects. Ammonium nitrogen concentration in the leachate from recirculation lysimeter 2 was always higher than that of recirculation lysimeter 1, confirming that the higher recirculation rate and volume of liquid phase, the more effective flushing of leachate through the waste mass.

In the two hybrid lysimeters a significant decrease in ammonium nitrogen concentrations from about 700 down to less than 2 mg/kg DM was observed. A similar tendency has been previously observed by Ritzkowski et al. (2006), Erses et al. (2008), and Long et al. (2009) when they studied the effect of aeration on nitrogen removal from old MSW.

The downtrends for ammonium nitrogen concentration in the leachates from two hybrid lysimeters 3 and 4 are similar. However, a dramatical slope for ammonium nitrogen concentration of the hybrid lysimeter 4 with higher aeration rate and time was observed about 10 days prior to that of the hybrid lysimeter 3, showing that increased aeration rate and/or frequency might have effectively triggered the microbial bacteria responsible for the oxidation of ammonium.

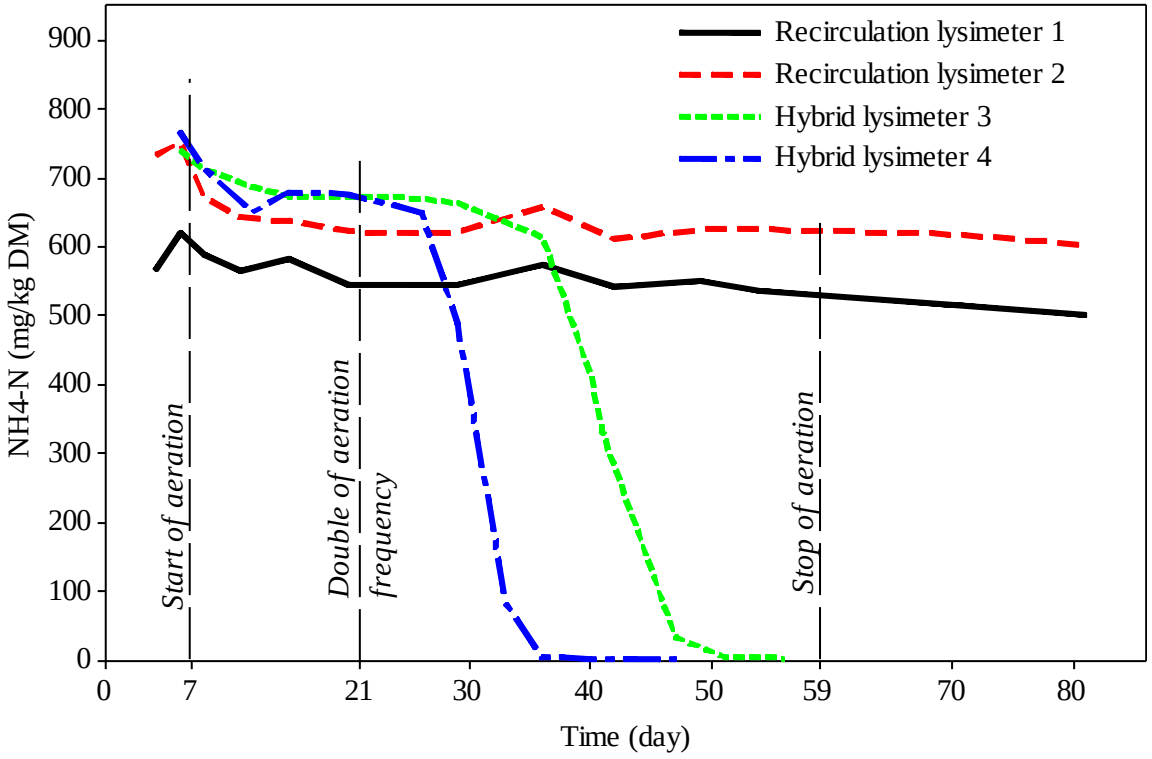


Figure 5.1. Ammonium nitrogen concentrations in the leachates

These declines in ammonium nitrogen concentrations were followed by rapid increases then decreases in nitrite and nitrate nitrogen concentrations (Figures 5.2 and 5.3), confirming that nitrification and subsequent denitrification have occurred.

5.2.2 Nitrite and nitrate nitrogen

During the increase of nitrite and nitrate nitrogen in the leachates from two hybrid lysimeters, concentrations of nitrite nitrogen in leachate in most cases were higher (around 4 to 5 times) than that of nitrate nitrogen (Figures 5.2 and 5.3).

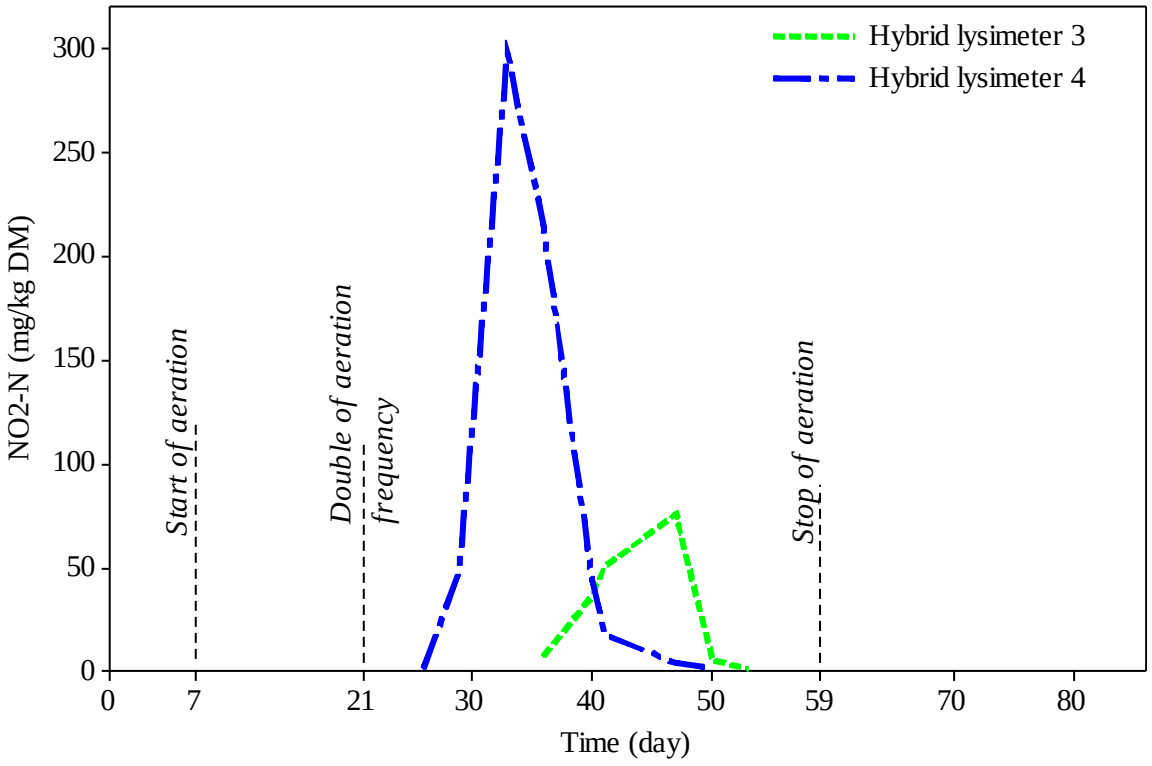


Figure 5.2. Nitrite nitrogen concentrations in the leachates

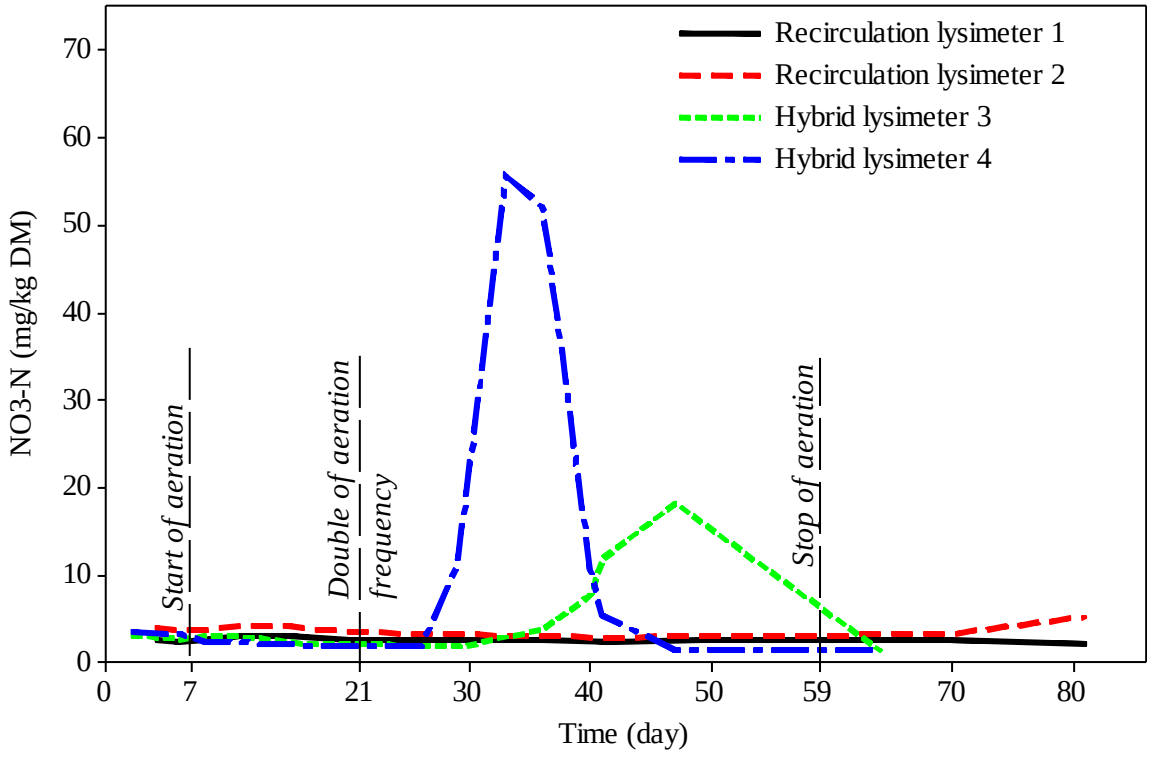


Figure 5.3. Nitrate nitrogen concentrations in the leachates

This effect could be attributed to either single or a combination of two following possibilities:

(i) The conversion rate of ammonium into nitrite was higher than that of nitrite into nitrate;
(ii) The denitrification process has occurred in the non-aerating time during the intermittent process, which converted nitrate into nitrogen-containing gases.

The latter means that the nitrification and denitrification were alternately underway during the intermittent aeration.

After reaching maximum values, nitrite and nitrate nitrogen concentrations in the leachates from two hybrid lysimeters rapidly decreased to very low levels. This is a sound evidence of the denitrification process.

The increases of nitrite and nitrate nitrogen concentrations in the leachate from the hybrid lysimeter 4 also appeared approximately 10 days before those of the hybrid lysimeter 3. Such variations once again indicate that the higher aeration rate and frequency applied might have enhanced not only on the abovementioned ammonium oxidation, but also on the nitrifying bacteria performing the conversion of nitrite to nitrate.

Nitrate nitrogen concentrations in the leachates generated from two recirculation lysimeters were relatively small and seemed to be at stable levels, whereas nitrite nitrogen concentrations were under the detection limit of analytical method during the whole experimental period.

These results are totally different from those obtained by Raga and Cossu (2013) as they studied the effects of aerobic conditions at different temperatures on waste and leachate quality. They found no nitrite and nitrate in the leachate. However, the waste they used for the tests was old MSW and their tests were run under different boundary conditions as well.

5.2.3 Total Kjeldahl nitrogen

The values for concentration of TKN in the leachates from four lysimeters are shown and compared to the respective ammonium nitrogen values in Figure 5.4. It is noticed that TN was determined instead of TKN for the last two analytical events (days 70 and 81) in case of recirculation lysimeters 1 and 2.

The ratio of ammonium nitrogen on TKN ranges from 89.3 - 98.2 %; 87.0 - 92.6 %; 62.5 - 92.7 %; and 62.1 - 94.8 % for LSR 1; 2; 3; and 4, respectively. As far as the

intermittent aeration progressed, such ratio was getting lower (62.5 % on day 47 for the hybrid LSR 3 and 62.1 % on day 40 for the hybrid LSR 4). This indicates that the conversion degree of organic nitrogen compounds decreasingly occurred during the aeration process. In other words, the remaining organic nitrogen substances at the end of the aeration should be refractory ones.

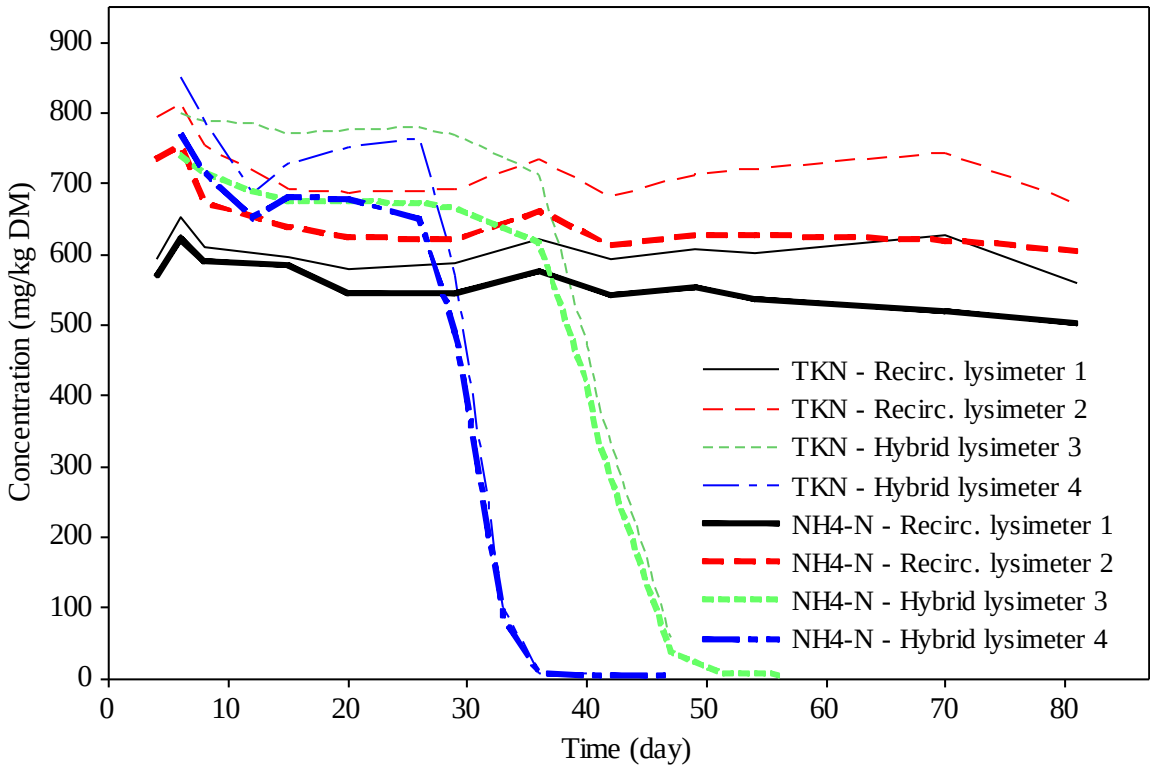


Figure 5.4. Concentration of TKN and NH_4-N in the leachates from four small lysimeters

5.2.4 Evaluation of variations in nitrogen compounds

The significant changes in ammonium, nitrite, and nitrate nitrogen as well as TKN (or TN) concentrations did not occur straight away after the application of aeration. After doubling the aeration frequency, a lag phase of approximately two weeks and one week regarding the hybrid lysimeters 3 and 4, respectively, can be observed. Processes leading to this phenomenon will be investigated in further studies.

During the experimental period the pH values for two recirculation lysimeters were relatively steady in the range from around 6.9 to 7.3; meanwhile, these values in the case of two hybrid lysimeters increased from about 6.7 to 7.8 during the first four weeks of the test run, then had relatively stable values around 7.1 (Figure 5.5).

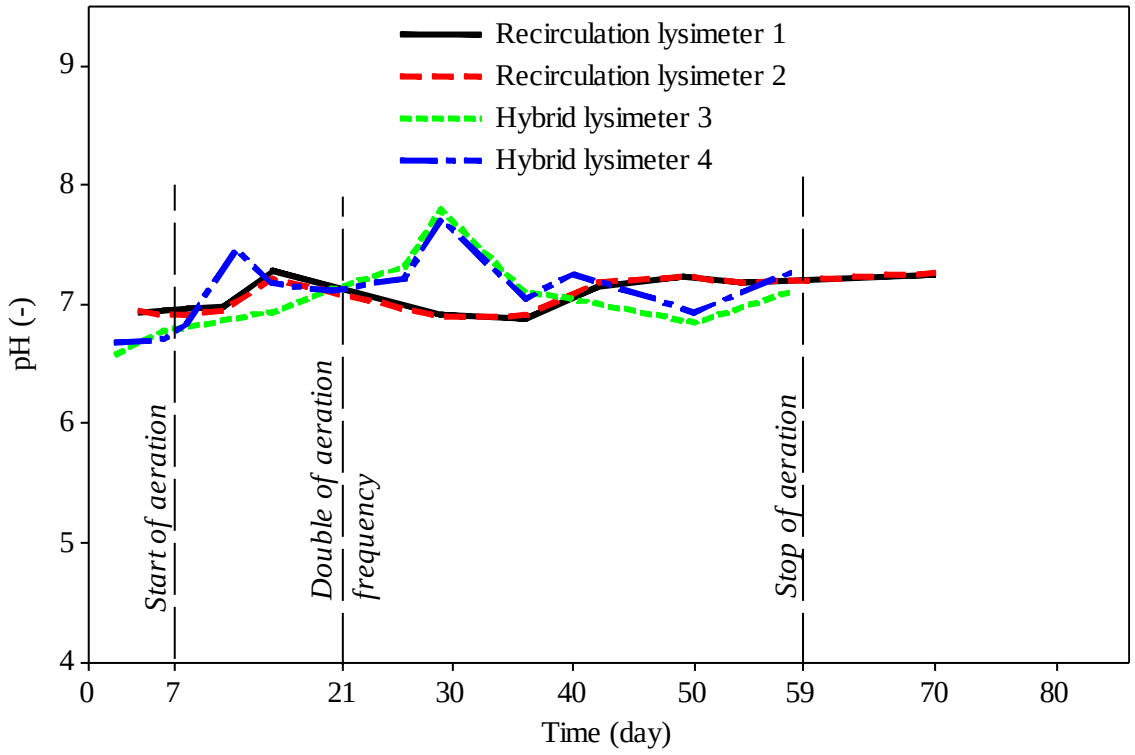


Figure 5.5. pH values in the leachates

It is well known that the volatilisation of ammonia gas only starts especially at pH values above 8 and increasing temperature. Hence, it could be figured out that the volatilisation of ammoniac via gaseous phase was very low because of the pH ranges shown above and the operating temperature around 20 ^{o}C of the lysimeters. Nitrification and denitrification should be then the main reason for the changes in ammonium, nitrite, nitrate nitrogen as well as TKN (or TN) concentrations regarding the two hybrid lysimeters.

The analyses of the leachates from all four lysimeters showed that TKN concentrations were in most cases slightly higher than those of ammonium nitrogen (see Figure 5.4 above). These mean that the proportion of organic nitrogen is small. Regarding the two hybrid lysimeters 3 and 4, TKN concentrations were considerably reduced during the intermittent aeration process. Extremely low concentrations of TKN, nitrite and nitrate around day 47 (for the hybrid lysimeter 4) and day 60 (in case of the hybrid lysimeter 3) suggest that certain nitrogen-containing gases were emitted and lost.

5.3 Organic carbon parameters

5.3.1 Total organic carbon and chemical oxygen demand

Concentrations of TOC and COD in leachates (Figures 5.6 and 5.7) were from the very start much lower compared with those normally found in leachates from MSW landfills.

The reason is that most of easily degradable organic carbon compounds are removed during MBT process. TOC and COD in the leachates from the MBT lysimeters in this case should mainly include the moderately and hardly degradable organic compounds.

Figure 5.6 shows the exemplary for TOC that the concentrations of organic compounds slightly decrease during the aeration period. Meanwhile, Figure 5.7 shows a comparatively strong decline in COD values.

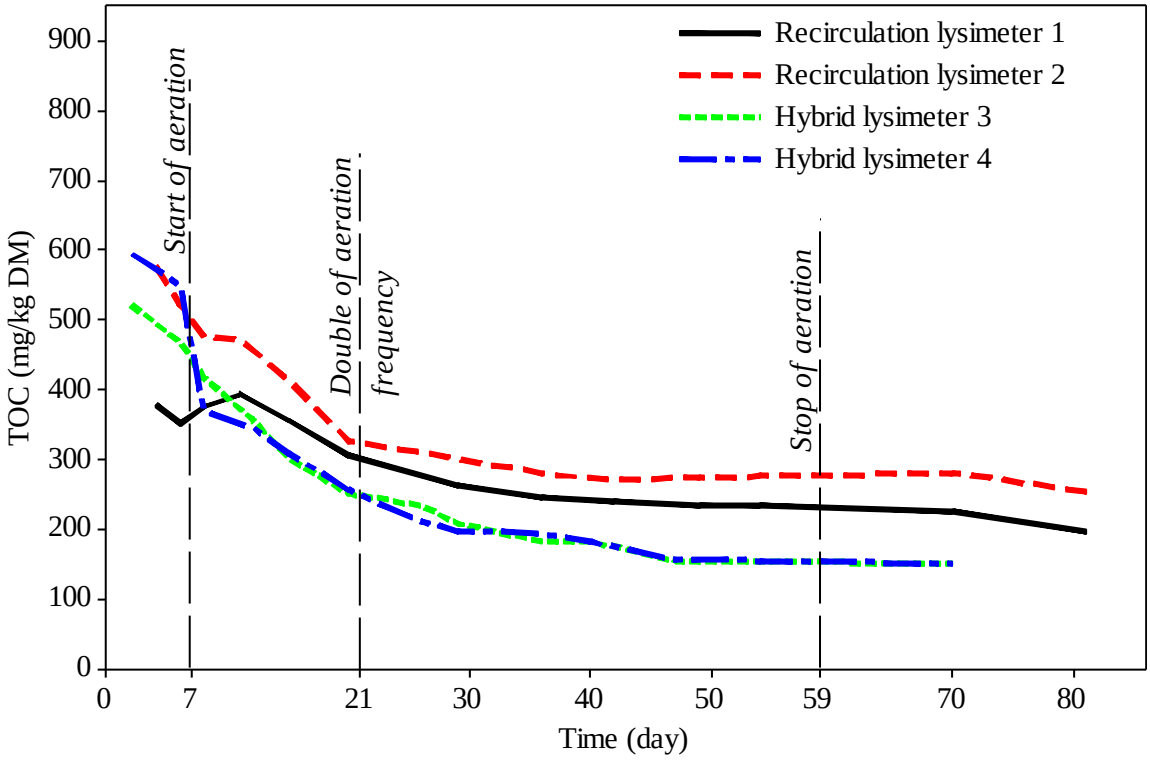


Figure 5.6. TOC concentrations in the leachates

The differences in aeration rate and time had little influence on the downtrend of TOC and COD, indicating that the application of intermittent aeration is less effective on the removal of organic carbon matter as compared with that of nitrogen compounds. At the later stage of the aeration process, the removal of organic carbon matters became less effective. After stopping the aeration, TOC and COD concentrations were almost stable at about 150 and 600 mg/kg DM, respectively. Their tendencies were similar to those for the recirculation lysimeters. The COD value of 600 mg/kg DM in this case should primarily comprise humic and fulvic acids.

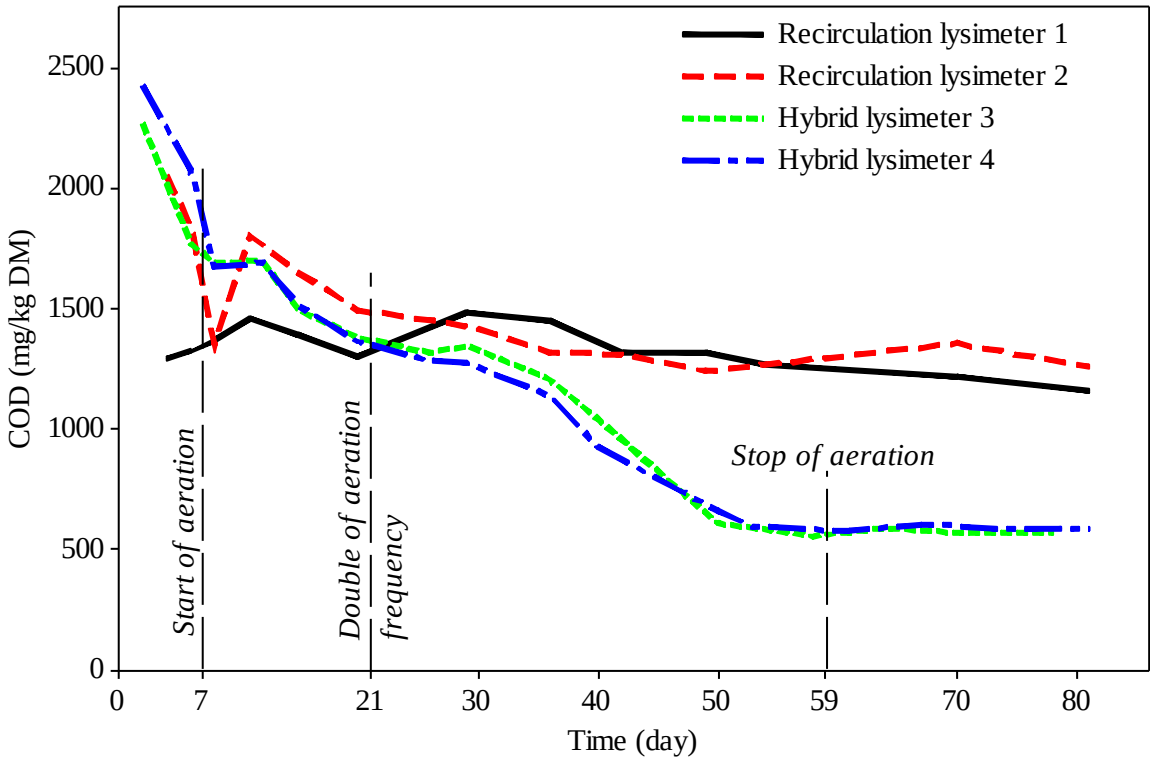


Figure 5.7. COD concentrations in the leachates

5.3.2 Biodegradability RI_4

RI_4 values of the MBT residue before loading and after unloading the lysimeters clearly showed the positive effect of intermittent aeration on the biodegradability of the loaded residue (Table 5.1).

Table 5.1. Effect of intermittent aeration on the biodegradability of loaded material

Parameter	**Unit**	**Input value**	**Output value**			
			Recirculation lysimeter		***Hybrid lysimeter***	
			LSR 1	**LSR 2**	**LSR 3**	**LSR 4**
RI_4	mg O_2/g DM	5.93	3.49	3.03	0.38	0.40

The already low RI_4 values of the input material is reduced by about 90 % to the very low values of 0.38 and 0.40 mg O_2/g DM in the hybrid lysimeters 3 and 4, respectively. The decreases in RI_4 for the recirculation lysimeters (from 5.93 to 3.49 and 3.03 for lysimeters 1 and 2, respectively) also show that biodegradable matters in the loaded waste have been

considerably flushed out to the leachate by the recirculation, but at a lower efficiency compared to the hybrid lysimeters.

Similar downtrends in TOC, COD, and RI_4 values were also reported by other authors as they investigated the effect of aeration on old MSW (Prantl et al., 2006; Ritzkowski et al., 2006; Raga and Cossu, 2013).

5.4 Nitrogen, TOC and COD removal efficiencies

Nitrogen, TOC and COD removal efficiencies were evaluated by comparing concentrations of TN, TOC and COD concentrations of the leachate at the beginning and the end of the lysimeters' operation.

The experimental results showed approximately 99 % nitrogen removal for the hybrid lysimeters, whereas the values are nearly 6 % and about 15 % for recirculation lysimeters 1 and 2, respectively (Figure 5.8).

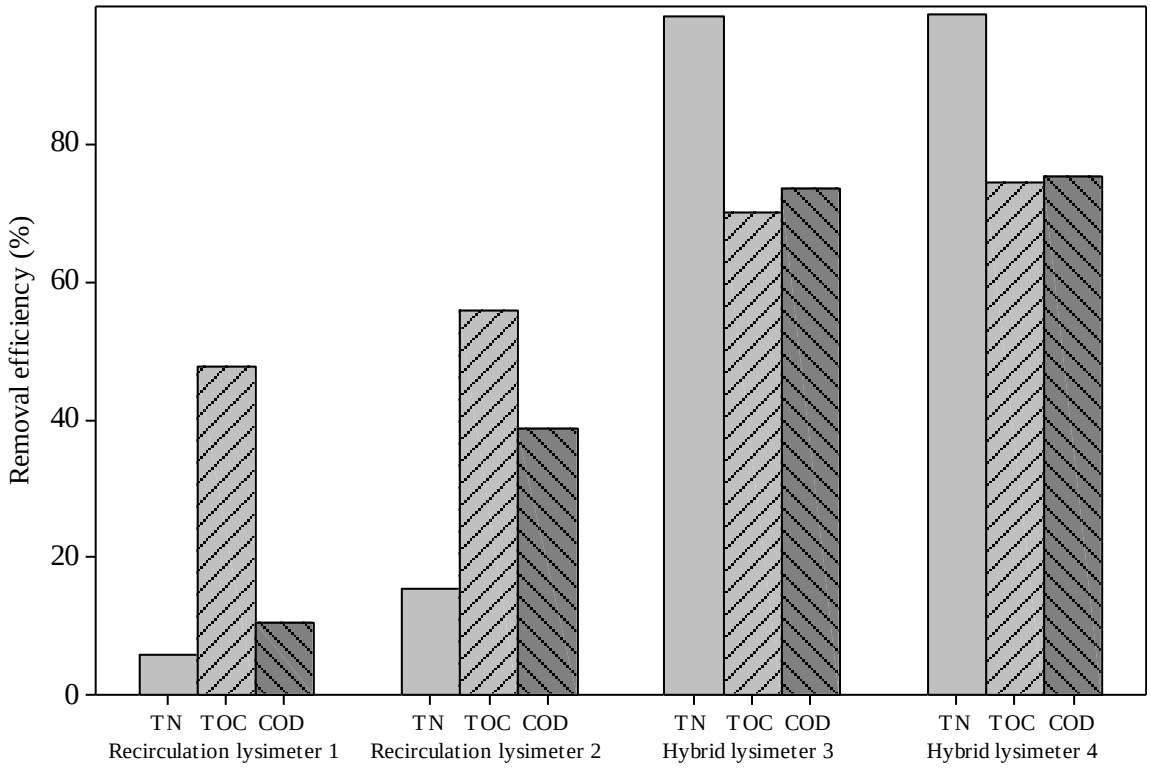


Figure 5.8. TN, TOC and COD removal efficiencies

It is also evident that the removal of organic carbon compounds can be achieved at certain levels for both cases of operation. However, the removal efficiency for the hybrid lysimeters is always higher than that of the recirculation ones.

The very high nitrogen removal efficiency in the two hybrid lysimeters can be attributed to the fact that nitrification and denitrification processes were almost completed. This is a special effect caused by the small scale of the lysimeters and the small mass of waste to be treated. Additionally, the more homogenous air distribution in the waste enhances the completion of the processes.

It must be taken into account for the test lysimeters that the mass of residue being loaded into each lysimeter is rather small; and moreover, the loading density is lower than the one which can be achieved on real MBT landfills. These effects result in a relatively fast conversion and reduction of nitrogen and biodegradable organic compounds in the loaded waste and its leachate. By the compaction of MBT residue during emplacement the pore volume available for gas and liquid transport is reduced to a minimum. The effect of conversion and reduction is therefore limited to a relatively small area nearby the place of aeration and/or recirculation. In the MBT landfills, porous niches and vacant spaces inside the landfill bodies should be less than those in the MBT residues before landfilling due to higher densities of deposited material. Thus, on one hand the diffusion of air in the porous system is more difficult by in-situ aeration, and on the other hand the leachate percolation rate through the high depth of highly compacted waste is smaller, which both lead to slower conversions.

When larger scale lysimeters are used, it is to be expected that removal efficiencies will be lower and the removal process will take longer time. Under the conditions of MBT landfill operation, a very irregular air distribution with zones of high oxygen-saturation and zones with nearly no oxygen are to be expected. The movement of oxygen between these zones is controlled by diffusion, so that removal efficiencies will be lower and also the removal progress will last longer.

The study results of Hrad et al. (2013) on the effect of lab- and full-scale in-situ aeration on the long-term emission of old MSW and old MSW landfill (where the old MSW was sampled for the lab-scale tests) stated that the effect of full-scale aeration was much less than that of lab-scale.

5.5 Summary

The combination technique of leachate recirculation and aeration has been applied for the first time to MBT residues in order to reduce organic carbon and nitrogen emissions after disposal on landfills. The results from the lab-scale lysimeter tests show a fast and significant decrease in ammonium and TKN, followed by a considerably rapid increase then decrease in nitrate and nitrite concentrations. These changing behaviours imply that nitrification occurred in the system, followed by denitrification.

The experimental results also indicate that a higher aeration rate and/or frequency have led faster to nitrogen conversion processes. Certain nitrogen-containing gases were most likely emitted from the system, which have to be analysed in future work. At the end of the tests, nitrogen removal efficiency of around 99 % for the hybrid lysimeters and of just about 6 to 15 % for the recirculation ones was determined.

The application of such combination technique also shows a moderate reduction in TOC and a relatively fast decrease in COD concentrations in the leachate from the hybrid lysimeters. However, the reduction effect is not as strong as for nitrogen compounds and the intermittent aeration seems to be effective only at the first stage of aeration. The aeration intensity has only little influence on the decrease of TOC and COD.

In case of the recirculation lysimeters, the slight decrease in concentrations of ammonium, TKN, TOC, COD in their leachate, as well as RI_4 values should be mainly caused by flushing effects.

It is to be expected that the results of the small lab-scale (1.34-L) lysimeter tests were influenced by the small amount of MBT residue investigated and the observation time after stopping aeration. MBT residue is smaller in size and more homogeneous compared to fresh MSW, so that smaller lysimeters can be used. However, a larger residue mass always gives a better reflection of the waste composition.

In conclusion, the combination of intermittent aeration with leachate recirculation allows significant reductions in concentrations of nitrogen compounds and relative small reductions in organic matters in the MBT residues and the leachates generated compared with that of only leachate recirculation. The long-term efforts, costs and duration of leachate treatment in the aftercare phase can therefore be reduced considerably.

CHAPTER 6: INFLUENCE OF LEACHATE RECIRCULATION IN COMBINATION WITH AERATION ON THE QUALITY OF LEACHATE FROM MBT AND MSW MATERIALS - A STUDY IN LARGER LYSIMETERS

6.1 Introduction

In this chapter, the analytical results for the leachate regarding phase 3 are presented and discussed. The changes in boundary conditions for six lysimeters of phase 3 have been already described in section 3.3.3. However, it is necessary to mention such changing events again to ease the interpretation and discussion of the achieved experimental results.

Table 6.1 summarises the changes that have been applied to each lysimeter. The absence of lysimeter 4 in the figure is because of the fact that it was run under anaerobic condition for the whole experimental period.

Table 6.1. Operational diary and changes in aeration regime

Time (day)	**Changing event**
0	Loading material and installing aeration system for lysimeters 1, 2, 5, & 6; Installing aeration system for lysimeter 3
5	Start of irrigation and leachate recirculation for lysimeters 5 & 6
11	Start of irrigation and leachate recirculation for lysimeters 1, 2, 3, & 4
27	Start aerating lysimeters 2 & 3 intermittently
96	Start aerating lysimeter 1 continuously
111	Stop of aeration for lysimeter 2
117	Start aerating solid phase of lysimeters 5 & 6 intermittently
122	Start aerating liquid phase of lysimeters 5 & 6 intermittently
133	Stop of aeration for lysimeter 1; Shift to continuous aeration for lysimeter 3
168	Increase aeration rate for the liquid phase of lysimeter 3
200	Increase aeration rate for the solid phase of lysimeters 5 & 6
265	Shift back to intermittent aeration for lysimeter 3
310	End of operation for lysimeters 5 & 6
413	End of operation for lysimeters 1, 2, 3, & 4

Excluding the changing events for five aerated lysimeters, leachate recirculation has been deployed to all six lysimters throughout the experimental time.

In terms of loading materials, the six lysimeters can be devided into three groups, in which two lysimeters 1 and 2 loaded with MBT residue create group 1. Group 2 includes lysimeters 3 and 4 that contained an old MSW material. The MSW material used for these two lysimeters was actually undergone the anaerobic condition and leachate recirculation in phase 1 for one year. Two lysimeters 5 and 6 were loaded with fresh MSW, assigning as group 3.

6.2 pH value, electrical condunctivity and temperature

Figure 6.1 shows the curve of pH values over time. The pH values for the MBT lysimeters 1 and 2 were in the range of about 6.5 to 7.6, and mostly fluctuated around 7. Such values are comparatively similar to those of the leachates generated from four small 1.34-L lysimeters in phase 2 (see Chapter 5).

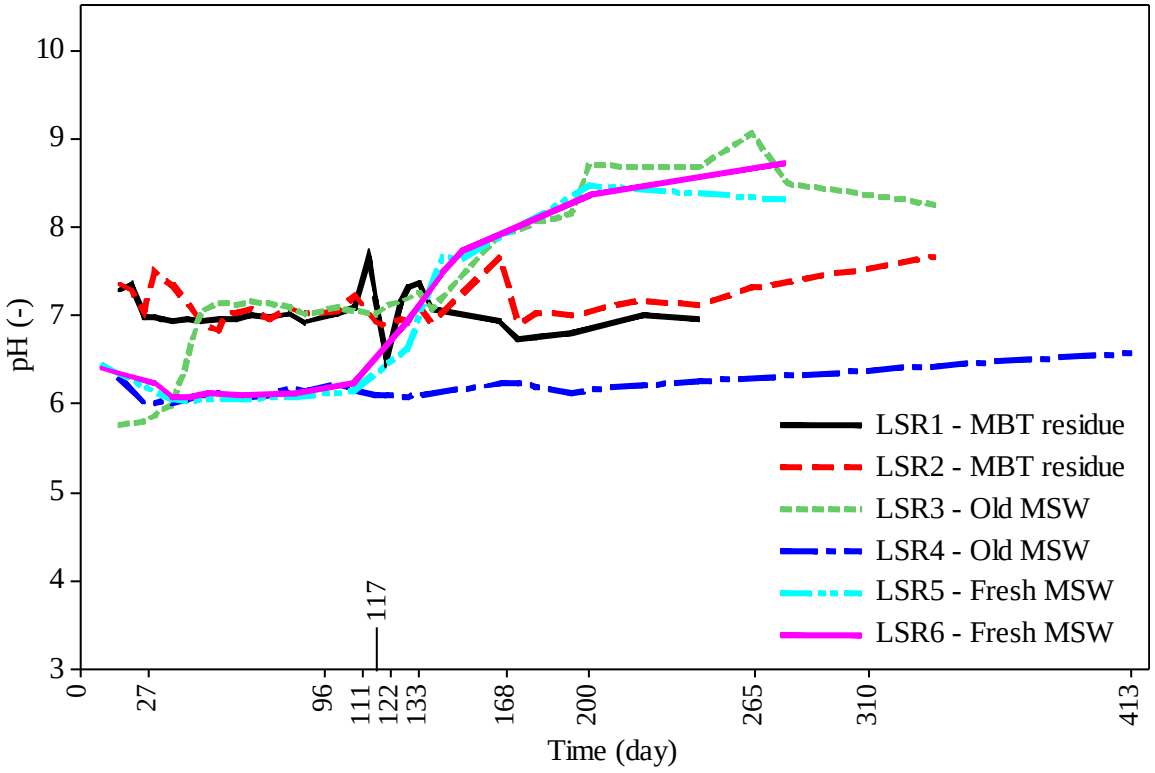


Figure 6.1. Variation in pH of the leachates

The leachates from the four other MSW lysimeters have initial pH values around 6. However, pH values started increasing as aeration regime progressed over lysimeters 3, 5, and 6. That can be attributed to the stripping of carbon dioxide by air regarding the aerated lysimeters. A decrease in carbon dioxide leads to a decrease in carbonic acid and

bicarbonate ion concentrations consuming hydrogen ions (Kim, 2005). The pH values of the leachates from the three aerated MSW lysimeters well agree with the literature data which have been reported to be in the range from 7.0 to 9.0 by several authors (Stessel and Murphy, 1992; O'Keefe and Chynoweth, 2000; Agdag and Sponza, 2004; Kim, 2005). In case of two MBT lysimeters, carbon dioxide formation as well as stripping should be very low due to the stabilised characteristics of the loaded material.

It can be observed that the highest pH values for lysimeters 3, 5, and 6 were 9.0, 8.5, and 8.7, respectively. Meanwhile, pH of the leachate from lysimeter 4 was stable at approximately 6.0 throughout the whole test period.

The change in EC of the leachates is given in Figure 6.2. It can be clearly seen that aeration had little effect on the reduction of EC values for the aerated MBT lysimeters, whereas there is a strong descending trend for the EC values of the leachates from the three MSW aerated lysimeters.

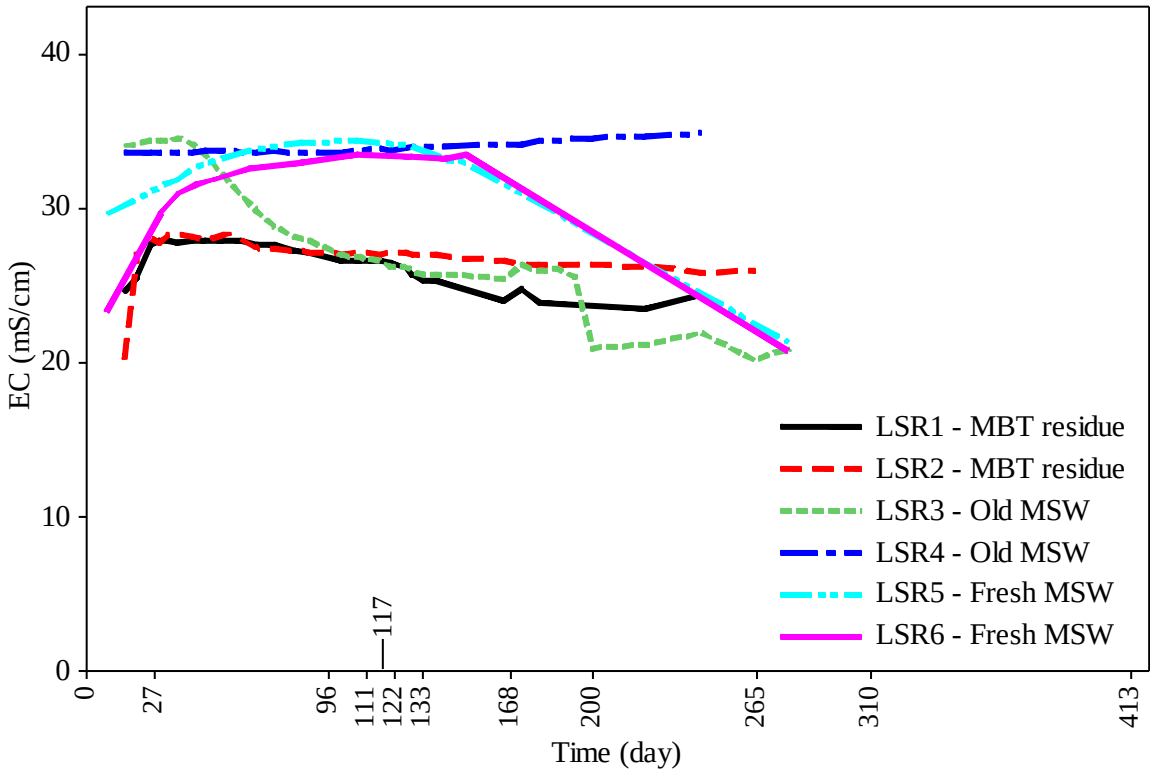


Figure 6.2. Variation in EC of the leachates

Significant decrease in EC values of the leachate mainly occurs due to the dilution effect. This effect has been clearly observed regarding two MBT lysimeters 1 and 2 in phase 1 which was previously mentioned in Chapter 4. However, the dilution effect in case of the lysimeters in phase 3 should be very small due to the large total volume of leachate in each lysimeter compared with a little volume of leachate compensation after every

sampling. The decrease in EC values of the leachates from aerated lysimeters may attribute to the reduction of sulphate concentration which will be discussed later in section 6.5.

The anaerobic MSW lysimeter 4 had a stable EC value of approximately 34 mS/cm for the whole test period, suggesting no change in the ion concentrations of the liquid phase.

Taking into account for temperature parameter, there was a little difference between the internal temperature and the external one in each lysimeter due to the regular recirculation of its leachate.

As for the two MSW lysimeters 5 and 6 in warm chamber, the internal temperature was normally 5 - 6 °C higher than the surrounding temperature. The internal and external temperature of the four other lysimeters and all lysimeters in phases 1 and 2 (operated under room temperature) fluctuated around 20 °C. Small amount of loading material and the regular leachate recirculation should have been the reason for such slight temperature difference between the inside and outside in case of two lysimeters 5 and 6.

In real conditions of MSW landfills that have no leachate recirculation, the huge amount and volume of landfilled wastes are less affected by the surrounding temperature. The temperature inside the landfill body may build up and reach approximately 65 - 70 °C in certain circumstances. Reasonable warm conditions certainly lead to faster decomposition and shorter time for stabilisation of the deposited wastes.

6.3 Nitrogen parameters

6.3.1 Ammonium nitrogen

Due to ammonification process, a part of the nitrogen pool is converted into ammonium ion. This was clearly observed in case of the four lysimeters 3, 4, 5, and 6 containing MSW (Figure 6.3).

Concentrations of ammonium nitrogen in the leachates from two lysimeters 5 and 6 containing fresh MSW have increased dramatically during the first 50 days since the beginning of their operation. Similar rising trends, but with lower slopes, have been also observed in two MSW lysimeters 3 and 4 as they underwent phase 1 (see Chapter 4). The much higher ammonification rate in case of two MSW lysimeters 5 and 6 (compared with that of two MSW lysimeters 3 and 4 in phase 1) clearly shows that warm temperature could enhance the degradation of the waste.

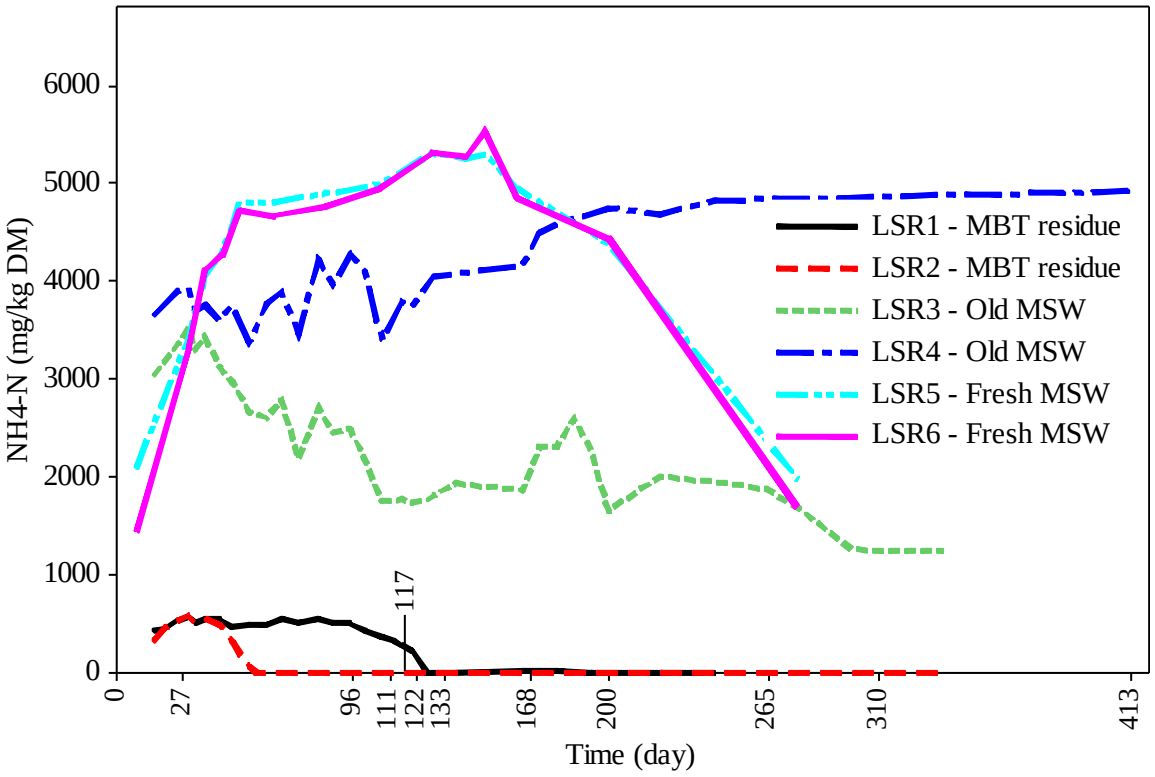


Figure 6.3. Variation in NH_4-N concentration of the leachates

Figure 6.3 shows that the concentration of ammonium nitrogen in the leachate from lysimeter 4 highly fluctuated for approximately 3 months since the start-up of phase 3. The fluctuation should be due to the uneven flushing and/or leaching as the leachate is drained through the lysimeter. This effect could derive from the fact that lysimeter 4 has been moved to another place before starting phase 3. Such movement of the lysimeter should have led to the shake and/or vibration of the internal loading material. The average ammonium concentration in the leachate regarding this fluctuated duration was high at approximately 3,800 mg/kg DM. At the end of the operation, ammonium nitrogen concentration was very high at about 4,600 mg/kg DM.

It is also clearly seen that the maximum ammonium nitrogen concentrations of the leachates from MSW lysimeters are about 7 - 10 times higher than those of the leachates from MBT lysimeters. It was observed that as soon as the aeration was applied to five lysimeters 1, 2, 3, 5 and 6 (day 96 for lysimeter 1; day 27 for lysimeter 2 and 3; day 117 and 122 for the solid and liquid phases, respectively in case of lysimeters 5 and 6), ammonium nitrogen concentrations in their leachates started to decrease.

Ammonium nitrogen concentrations in the leachates from lysimeters 1 and 2 were sharply dropped almost right after the introduction of continuous and intermittent aeration. It is observed that there was no lag phase in these lysimeters, whereas a lag

stage was present as applying intermittent aeration to the small 1.34-L MBT lysimeters in phase 2 (see Chapter 5).

Concentration of ammonium nitrogen in the leachate from lysimeter 3 appeared to have some rising points surrounding the time that the increase in aeration rate for its liquid phase was implemented (day 168). However, such increase should not have been due to dissimilatory nitrate reduction process (see Eqs. 2-12 and 2-13 in section 2.2.5) because of two reasons:

(i) The condition inside the lysimeter and its leachate container was almost aerobic due to the fact that it was continuously aerated during the time from day 133 to day 265. This duration also covers the time that some rising points appeared.
(ii) Nitrate nitrogen concentration in its leachate was not increased, but rather low and stable.

However, the exact reason for such unusual change has not yet been revealed so far.

6.3.2 Nitrite and nitrate nitrogen

Variations in nitrite and nitrate nitrogen concentrations of the leachates are shown in Figures 6.4 and 6.5, respectively.

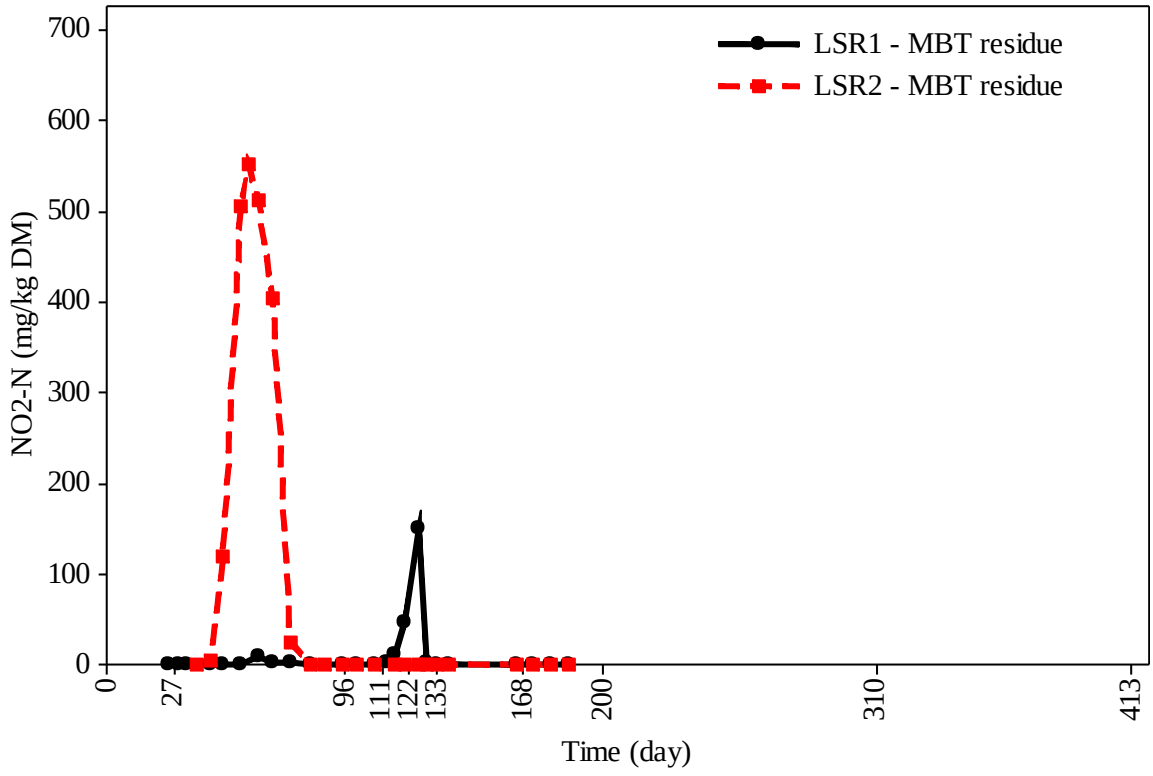


Figure 6.4. Variation in NO_2-N concentration of the leachates

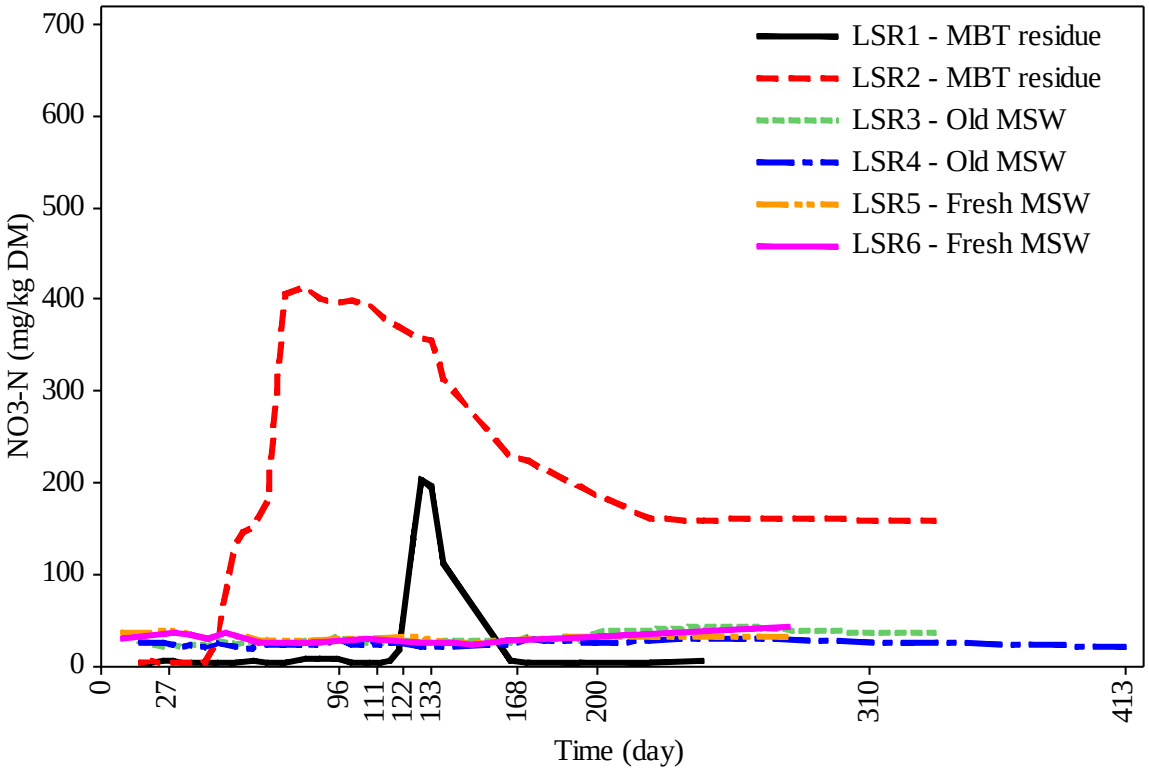


Figure 6.5. Variation in NO_3-N concentration of the leachates

It can be seen from Figures 6.4 and 6.5 that the major changes in nitrite and nitrate nitrogen concentrations occurred only in case of the two MBT lysimeters 1 and 2. Both nitrite and nitrate concentrations in their leachates dramatically increased as ammonium nitrogen values decreased. No nitrite nitrogen in either old MSW lysimeters or fresh MSW lysimeters was detected. Nitrite nitrogen concentrations of the leachates from lysimeters 1 and 2 were at highest values of 150 and 553 mg/kg DM, respectively. Furthermore, concentrations of nitrate nitrogen in such leachates peaked at 203 and 413 mg/kg DM, respectively. These values then rapidly dropped to very low level and finally under the detection limits of the analytical method (< 0.23 mg/L NO_2-N by using Hach Lange cuvette test kits LCK 339), excepting nitrate nitrogen concentration in the leachate from lysimeter 2.

The changing trends in nitrite and nitrate nitrogen, as well as ammonium nitrogen concentrations for two MBT lysimeters 1 and 2 are mostly similar to those for the two small 1.34-L hybrid MBT lysimeters in phase 2 (see Chapter 5). However, lysimeter 2 with a purposeful longer aeration time has showed a little difference in the downtrend of nitrate nitrogen concentration.

The lysimeter 2 was intentionally kept aerated until day 111 after the start-up of operation to observe the behaviour of nitrogen parameters. In fact, the concentrations of ammonium

nitrogen (3.5 mg/kg DM) and nitrite nitrogen (0.2 mg/kg DM) were at extremely low values on day 82. The concentration of nitrate nitrogen in its leachate was decreased from the maximum value of 413 mg/kg DM down to approximately 160 mg/kg DM and kept stabilising at this level until the end of the test. This fact can be attributed to the incompletion of denitrification, which is unexpected in regards to nitrogen removal purpose. However, such nitrate nitrogen concentration downtrend in case of lysimeter 2 could give a suggestion that the aeration should have quit as soon as ammonium and nitrite nitrogen concentrations of the leachate already reached low levels (i.e. around day 82 from the start-up of the operation). It is assumed that if the aeration was stopped at that point, anaerobic and/or anoxic conditions would be re-established to facilitate denitrification process. The maximum concentrations of ammonium nitrogen (582 mg/kg DM), nitrite nitrogen (553 mg/kg DM), and nitrate nitrogen (413 mg/kg DM) in its leachate clearly indicate a very high conversion yield in view of nitrification stoichiometry.

The MBT lysimeter 1 was undergone 37 days of continuous aeration to have extremely low levels of ammonium, nitrite, and nitrate nitrogen in its leachate. Meanwhile, the MBT lysimeter 2 was gone through 44 days under intermittent aeration, so that ammonium and nitrite nitrogen concentrations of its leachate were at very low levels. The peak values of nitrite and nitrate nitrogen in the leachate from lysimeter 2 in this phase are about 3.7 and 2 times, respectively higher than those from lysimeter 1. Meanwhile, the peak values for ammonium nitrogen concentrations of the leachate from both lysimeters 1 and 2 are of approximate identity. Such values imply that nitrification occurring in case of lysimeter 1 is at much lower performance in comparison to that of lysimeter 2.

As for four MSW lysimeters, nitrate nitrogen concentration in their leachates was relatively stable in the range from 20 - 40 mg/kg DM. However, nitrite nitrogen concentration was always under the detection limit either for anaerobic MSW lysimeter 4 or aerated MSW lysimeters 3, 5, and 6. This indicates that aeration has no influence on altering nitrite and nitrate nitrogen parameters. However, there were relatively rapid reductions of ammonium nitrogen concentrations in the leachates from three aerated MSW lysimeters as the aeration progressed.

6.3.3 Total nitrogen

Regarding TN parameter, the experimental results clearly show that the concentration curve of TN in the leachates from two MBT lysimeters only bears a resemblance to that of ammonium nitrogen prior to the introduction of aeration (by comparing Figures 6.6 and 6.3).

Remarkable variations in ammonium, nitrite and nitrate nitrogen values during the aeration have led to two different tendencies in TN.

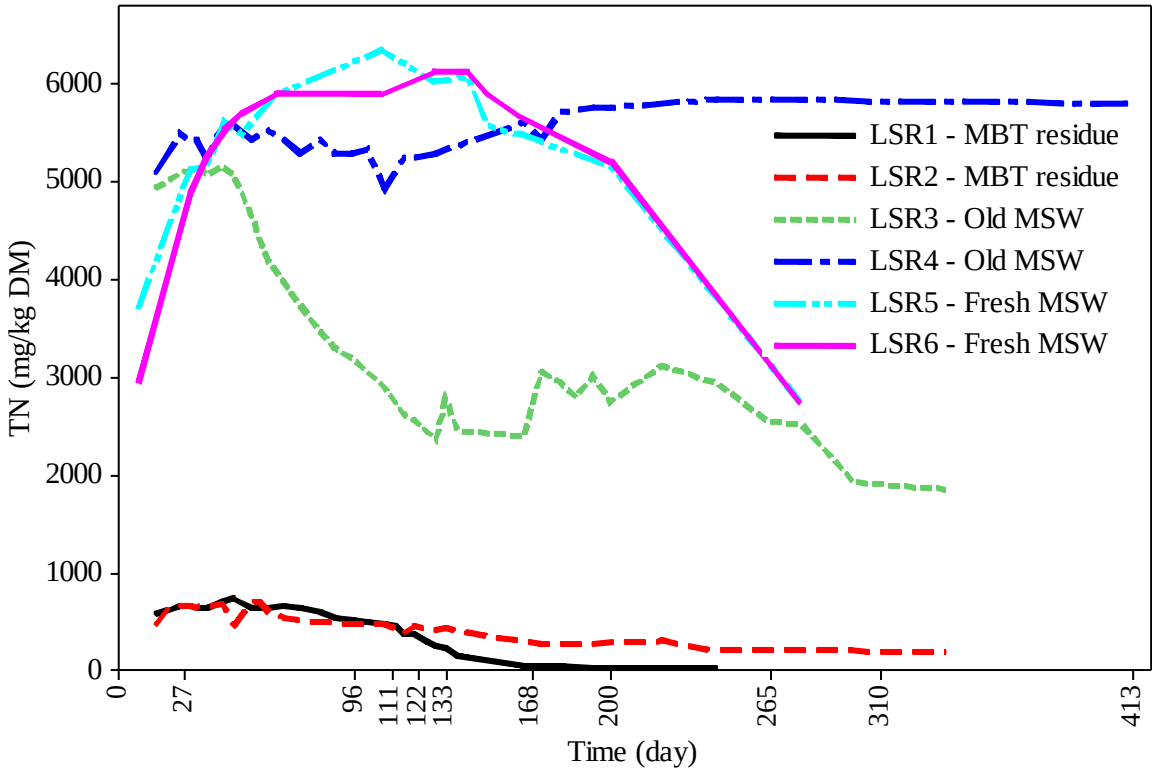


Figure 6.6. Variation in TN concentration of the leachates

TN values of the leachate from MBT lysimeter 1 have shown a steady decrease from around 500 mg/kg DM on day 96 (start of continuous aeration) to 229 mg/kg DM on day 133 (stop of the aeration) and reduced to about 11 mg/kg DM on day 243. In case of MBT lysimeter 2, the concentration of TN also decreased; however, it was less steady. TN values stabilised at around 200 mg/kg DM as nitrate nitrogen concentration in its leachate decreased to stable level of about 160 mg/kg DM at the latter stage of the aeration. This also means that organic nitrogen concentration in its leachate accounts for approximately 20 % of the TN value.

As for MSW lysimeters, TN changing trend and the corresponding tendency of ammonium nitrogen concentration in their leachates also resemble each other in shape, but, in a quantitative view, differ from each other to some certain extent. A combination of different aspects, including the reductions of ammonium nitrogen and TN values, the increasing pH trend, the stable values of nitrate nitrogen, as well as the undetected nitrite nitrogen suggests that the loss of nitrogen from three aerated MSW lysimeters should have been due to the volatilisation of ammonia gas. Nitrification should not have

happened to such MSW lysimeters regardless of either intermittent or continuous aeration that have been applied.

6.4 Organic carbon parameters

With respect to carbonaceous parameters, emission behaviours of the leachates from the lysimeters in this phase are discussed via the variations in concentrations of TOC, COD, and volatile fatty acids (VFA). The determination of VFA was not available for the leachates from lysimeters in phases 1 and 2. However, it is supplemented to phase 3 in order to facilitate the interpretation of the experimental results.

6.4.1 Variations in total organic carbon

Figure 6.7 shows the change in TOC concentration of the leachates from six lysimeters. The TOC concentration increased at the initial stage due to the hydrolysis of organics from the waste into the leachate. It can be also clearly seen that TOC values of the leachates from four MSW lysimeters are extremely high in comparison to those from two MBT lysimeters.

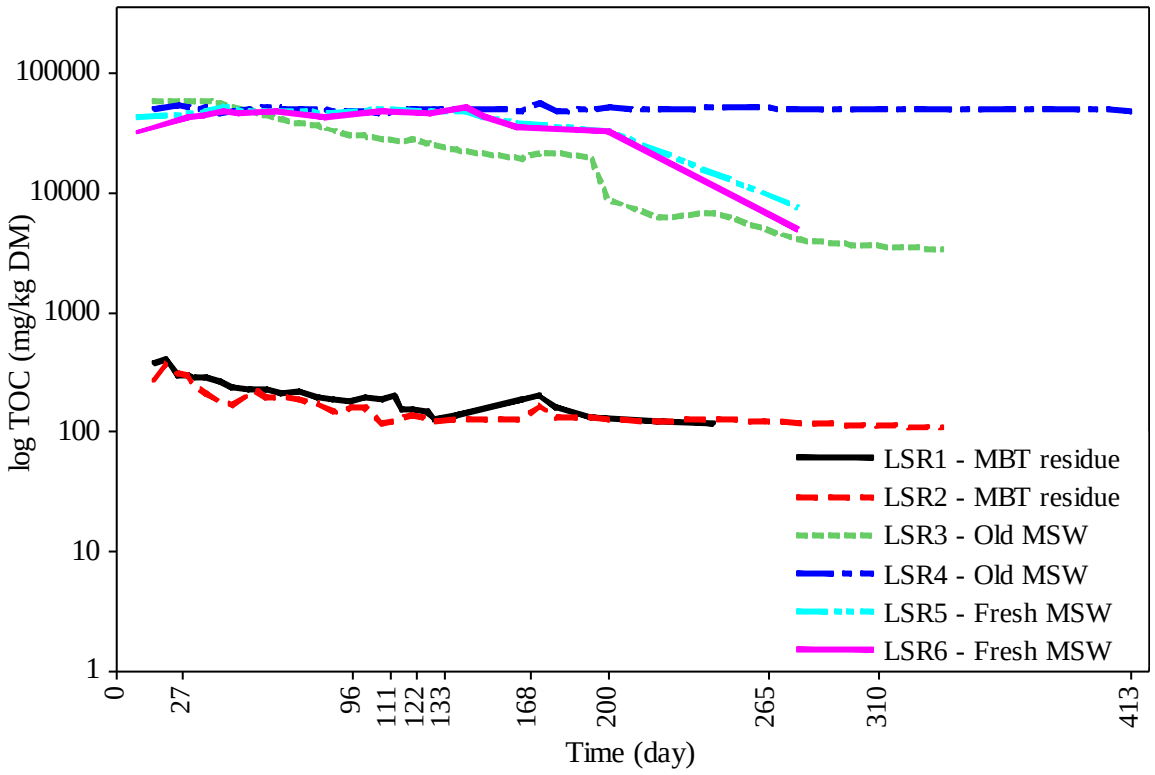


Figure 6.7. Variation in TOC of the leachates

The TOC values of the anaerobic MSW lysimeter 4 were stably fluctuated around 50,000 mg/kg DM. The maximum TOC values in the leachates from the MSW lysimeters 3, 5, and 6 were in the same magnitude order of about 51,000 - 58,500 mg/kg DM.

The TOC concentrations of the leachates from two MBT lysimeters 1 and 2 were few hundred times lower than those from four MSW lysimeters.

6.4.2 Variations in chemical oxygen demand

Variations in the concentrations of COD in the leachates from six lysimeters are illustrated in Figure 6.8.

COD values generally show similarity in shape, but at approximately 3.4 - 4 times higher with regard to TOC ones (by comparing Figures 6.8 and 6.7).

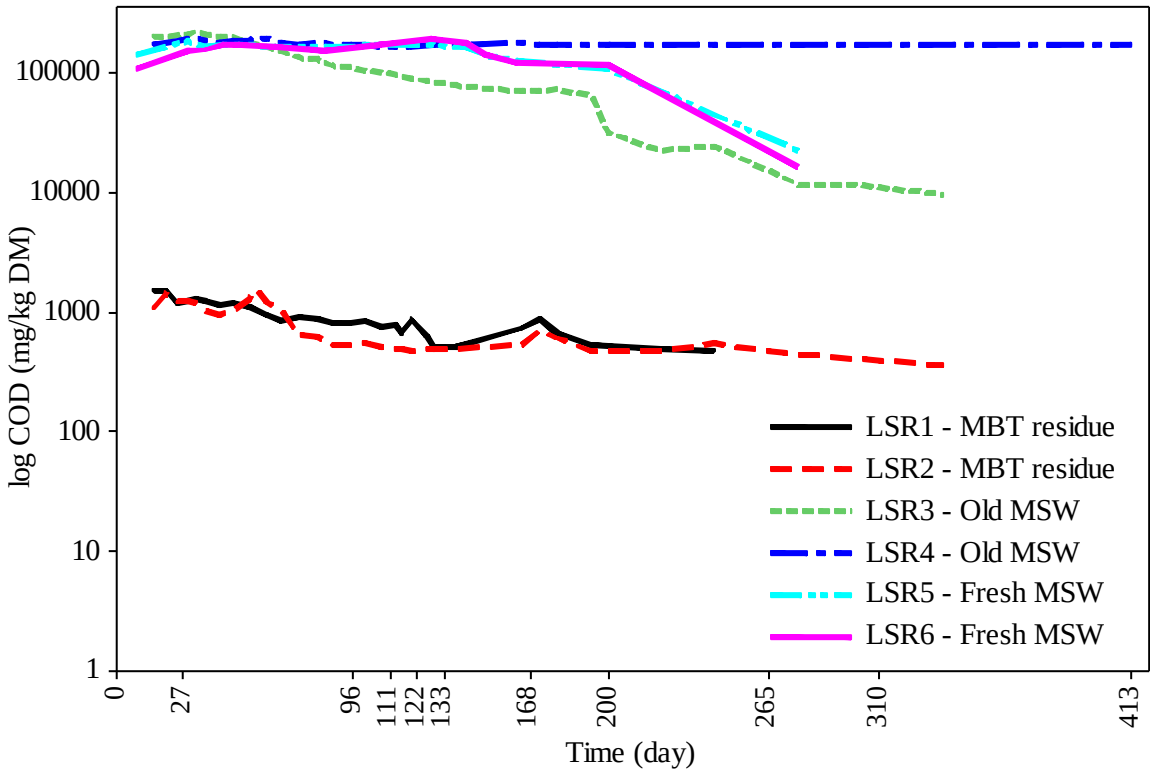


Figure 6.8. Variation in COD of the leachates

The COD values of the anaerobic MSW lysimeter 4 were stably fluctuated around 170,000 mg/kg DM. The maximum COD values in case of three MSW lysimeters 3, 5, and 6 were 217,265, 180,292, and 187,087 mg/kg DM, respectively. The COD concentrations of the leachates from two MBT lysimeters 1 and 2 were few hundred times lower than those from four MSW lysimeters.

As to be expected that aeration enables significantly faster biodegradation of organic matter, particularly in regard to three aerated MSW lysimeters 3, 5, and 6. The experimental results showed a similar trend to the findings of Cossu et al. (2003), who reported high COD values in the anaerobic lysimeter (around 20,000 mg/L) compared to the aerobic one (800 mg/L) after 120 days of operation. A study by Ritzkowski et al. (2006) stated a considerable decline of leachate COD concentrations after about 20 days of aeration. Raga and Cossu (2012) and Erses et al. (2008) also observed a strong reduction in COD concentrations of the leachates as they investigated the effect of aeration on old MSW.

It was very interesting that there were some increases in both COD and TOC concentrations during the aeration of two MBT lysimeters, and even though in case of the lysimeter 3 containing old MSW. Principally, an increase in COD during the start-up of nitrification process is an indication of nitrite build-up in the system. Though ammonium is not oxidised in the COD analysis, nitrite gets oxidised and hence conversion of ammonium to nitrite in the process results in an increase in the COD. However, the exact reasons for such increases are still unknown, and need further investigations in the future.

6.4.3 Variations in volatile fatty acids

The experimental results showed that total VFA concentrations in the leachates from two MBT lysimeters 1 and 2 were extremely low in comparison with those from MSW lysimeters (Figure 6.9).

The leachate from MSW lysimeter 4 has a very stable VFA concentration during the whole test duration, with an average value of 64,000 mg/kg DM which is about 700 times higher than that from two MBT lysimeters. Such values indicate that most of VFA has been successfully removed during the previous MBT process. The stable and high concentrations of VFA in the leachate from the MSW lysimeter 4 also imply that it did not get through the acidogenic phase. In this case, the acid concentrations (H_2CO_3 and VFA) should have exceeded the available alkalinity, MSW lysimeter 4 is considered to be “sour”, severely inhibiting the microbial activity, especially that of methanogens. High levels of VFA in the leachates from MSW lysimeters 3, 5, 6 (before aeration) and from lysimeter 4 result in low pH values which can bring about complete inhibition of both methane production and hydrolysis.

VFA concentrations in the leachate from lysimeter 1 fluctuated and had an average value of about 87.23 mg/kg DM. Lysimeter 2 generally shows a similarity to lysimeter 1, excepting some steep increases during the time from day 54 to day 67. At this period of

time, TOC and COD in the leachate from lysimeter 2 were also experienced some rising points.

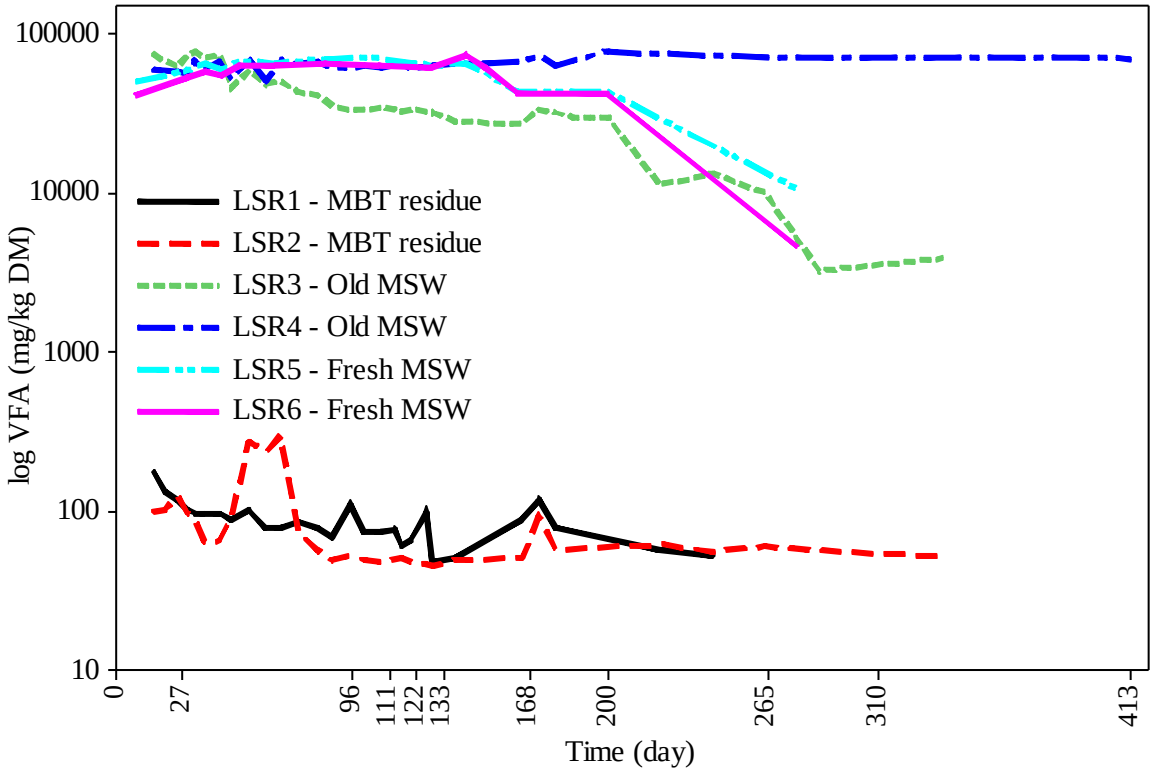


Figure 6.9. Variation in total volatile fatty acids of the leachates

6.5 Sulphate

The experimental results (Figure 6.10) showed that aeration had significant impact on the variations in SO_4^{2-} concentration of the leachates from aerated lysimeters.

It was observed for every aerated lysimeter (regardless of MBT or MSW) that SO_4^{2-} concentration rapidly increased nearly right after the implementation of aeration for a short time, then decreased.

The increase in SO_4^{2-} concentration should be due to the aerobic oxidation of reduced sulphur compounds (see Eqs. 2-17 and 2-18 in section 2.3.2.2).

The variations in nitrate and nitrite during the increase of sulphate imply that DEAMOX process (see Eq. 2-20 in section 2.3.2.2) should not have occurred in the system.

Figure 6.10 also shows, regarding two MBT lysimeters 1 and 2, that after the increase and then decrease in their SO_4^{2-} concentration, the SO_4^{2-} values were stabilised at around 1,700 and 1,600 mg/kg DM, respectively.

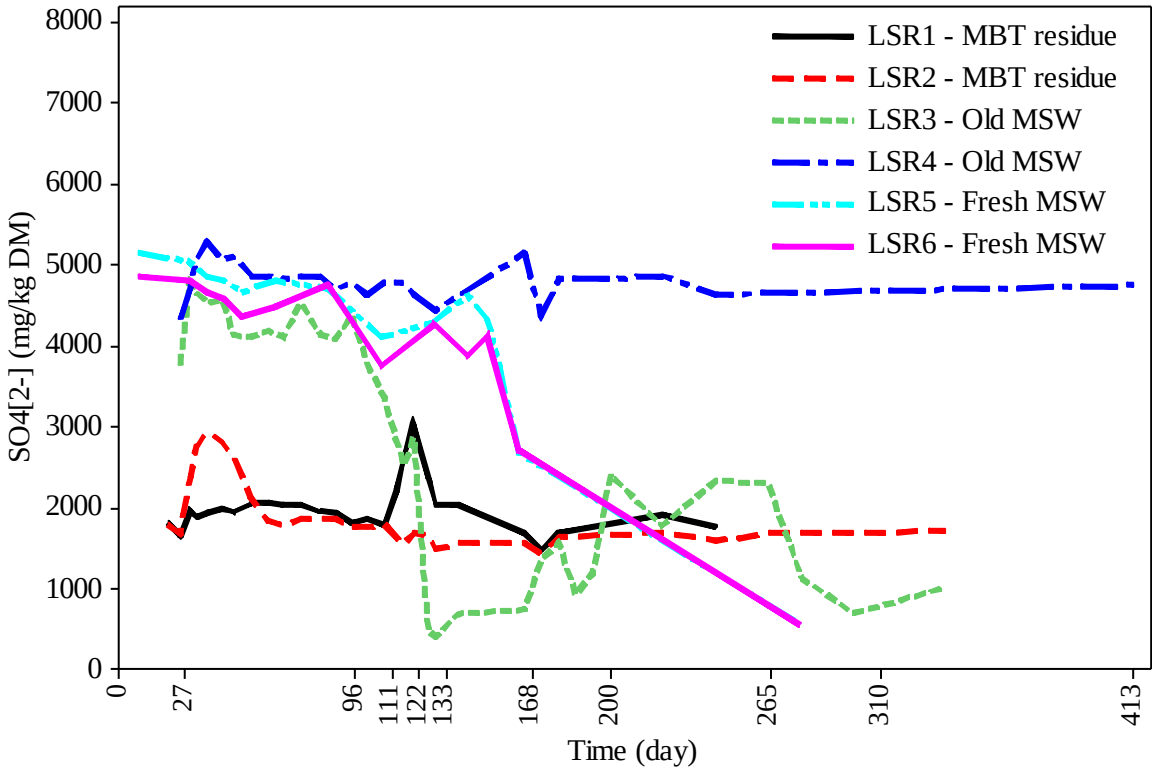


Figure 6.10. Variations in SO_4^{2-} of the leachates

Among three aerated MSW lysimeters 3, 5, and 6, two lysimeters 5 and 6 experienced a similar downtrend in SO_4^{2-} concentration, whereas the changing in this parameter in the leachate from lysimeter 3 was remarkably fluctuating. However, it can be recognised that the acts of changing intermittent aeration to continuous one (day 133) and increase in aeration rate for the leachate container of MSW lysimeter 3 (day 168) have caused the build-up again of sulphate. This behaviour can be explained again by the existence of reactions which were expressed via Eqs. 2-17 and 2-18 in section 2.3.2.2. The reduction of sulphate concentration is most likely owing to biological reduction (see Eq. 2-14 in section 2.3.2.2).

6.6 COD/SO_4^{2-} ratio

In considering the oxidation of organic compounds by sulphate reduction, it is well known that 1 g of sulphate can oxidise 0.67 g COD. Hence, for waste streams with a COD/SO_4^{2-} ratio of 0.67, there is in theory sufficient sulphate present to completely remove the organic matter present with sulphate reduction. For ratios exceeding 0.67, complete organic removal can only occur when methanogenesis occurs in addition to sulphate reduction (Lens et al., 1998).

Based on the experimental COD and SO_4^{2-} values, the ratio of COD/SO_4^{2-} is figured out.

It can be clearly seen from Figure 6.11 that the COD/SO_4^{2-} ratio is almost less than 0.67 regarding two MBT lysimeters.

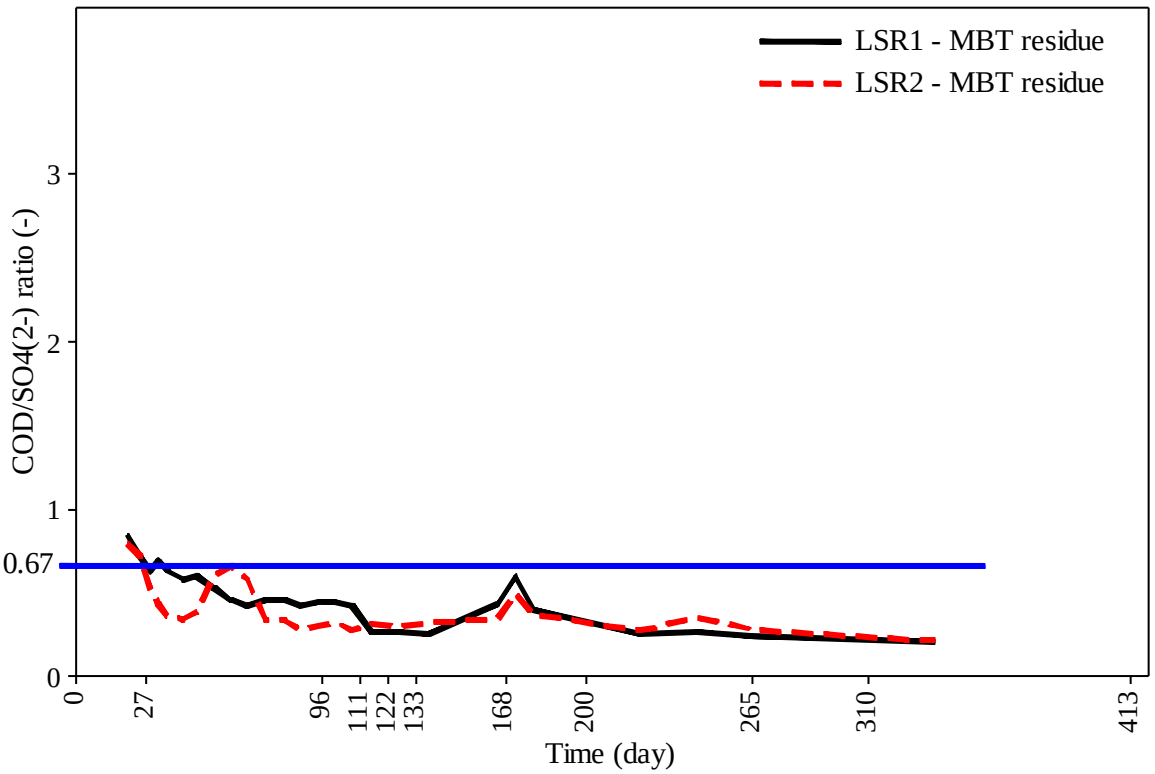


Figure 6.11. Ratio of COD/SO_4^{2-} for two MBT lysimeters

Meanwhile, it is much higher and slightly fluctuated around 36 for three MSW lysimeters 4, 5, and 6 (Figure 6.12). The stable value of COD/SO_4^{2-} in case of the MSW lysimeter 4 is because of the fact that not only COD and SO_4^{2-}, but also other concerned parameters from its leachate are stabilised throughout the whole testing operation.

As for two MSW lysimeters 5 and 6, the aeration brought to a strong influence on the reduction of COD and SO_4^{2-}, but the COD/SO_4^{2-} ratios were kept almost unchanged at 35.5 and 36.1, respectively. Such behaviour implies that COD and SO_4^{2-} decreasing rates should be similar.

The COD/SO_4^{2-} ratio in case of MSW lysimeter 3 was highly fluctuated in the range from 46.3 to 207.5 in the duration from day 126 to day 193 of the lysimeter operation. During this period, the leachate COD value from MSW lysimeter 3 has a regular downing trend; meanwhile the leachate SO_4^{2-} value was very much altered (it was 2,863.6, 460.0, 399.3, 678.1, 736.0, 1,361.7, 1,556.7, 918.2, and 1,190.5 mg/kg DM on day 120, 126,129, 138, 165, 172, 179, 186, and 193, respectively). The ratio was then stabilised at about 12 as both COD and SO_4^{2-} had a similar descending tendency.

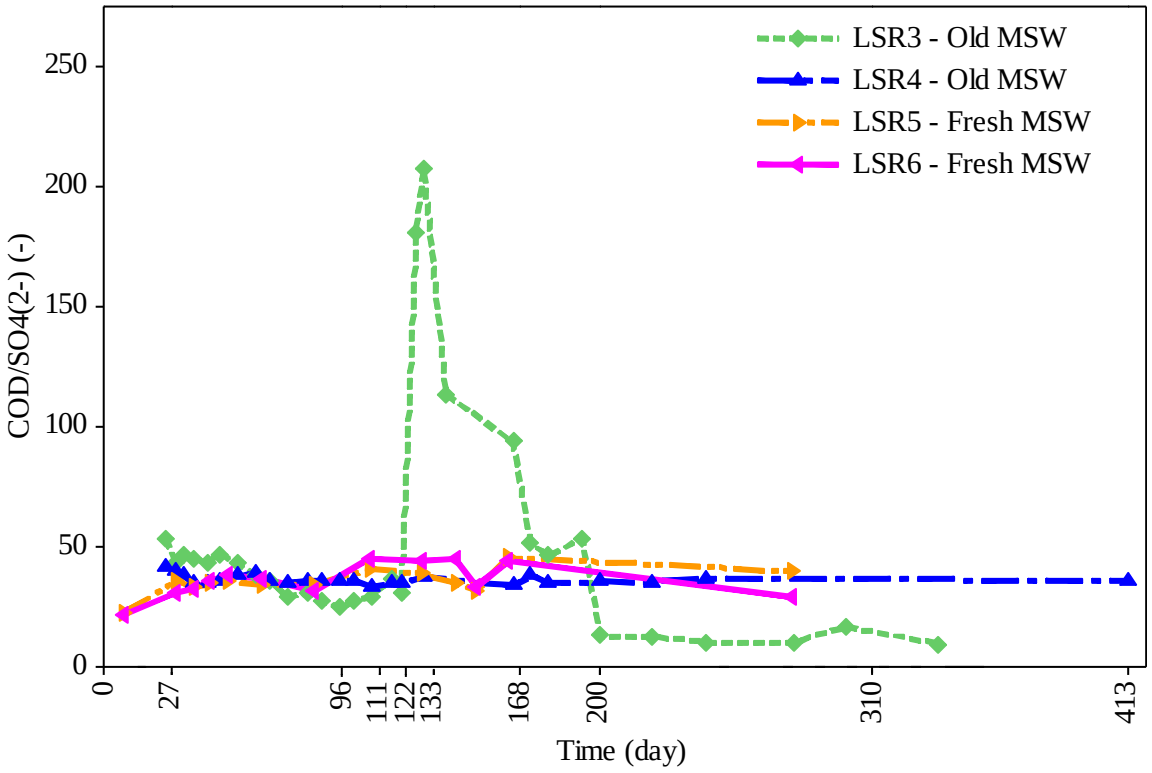


Figure 6.12. Variations in COD/SO_4^{2-} ratio for four MSW lysimeters

6.7 Biogas composition

The emitted gases from the test lysimeters were analysed whenever the gas bags were filled with a certain volume of gas. Biogas was measured for O_2, CO_2, and CH_4. Figures 6.13 - 6.18 depict the changes in composition of the biogas released from lysimeters 1 - 6 over a period of time, respectively.

The data from Figures 6.13 and 6.14 address that the exhausted gas from two MBT lysimeters as they are in anaerobic conditions contains nearly no methane and just a little amount of carbon dioxide. The low level of COD, TOC, and VFA in the leachates from two MBT lysimeters should be the reason for such behaviour of the biogas composition. It means that the methanogenic phase did not or very slightly occur in the MBT lysimeters.

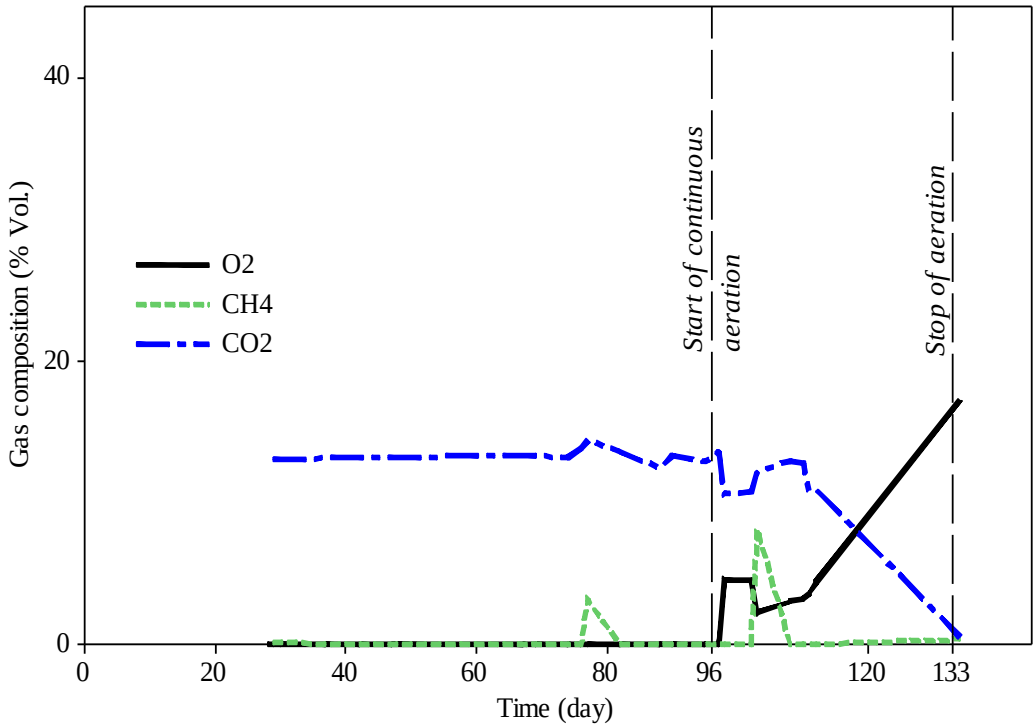


Figure 6.13. Variations in biogas composition from LSR 1

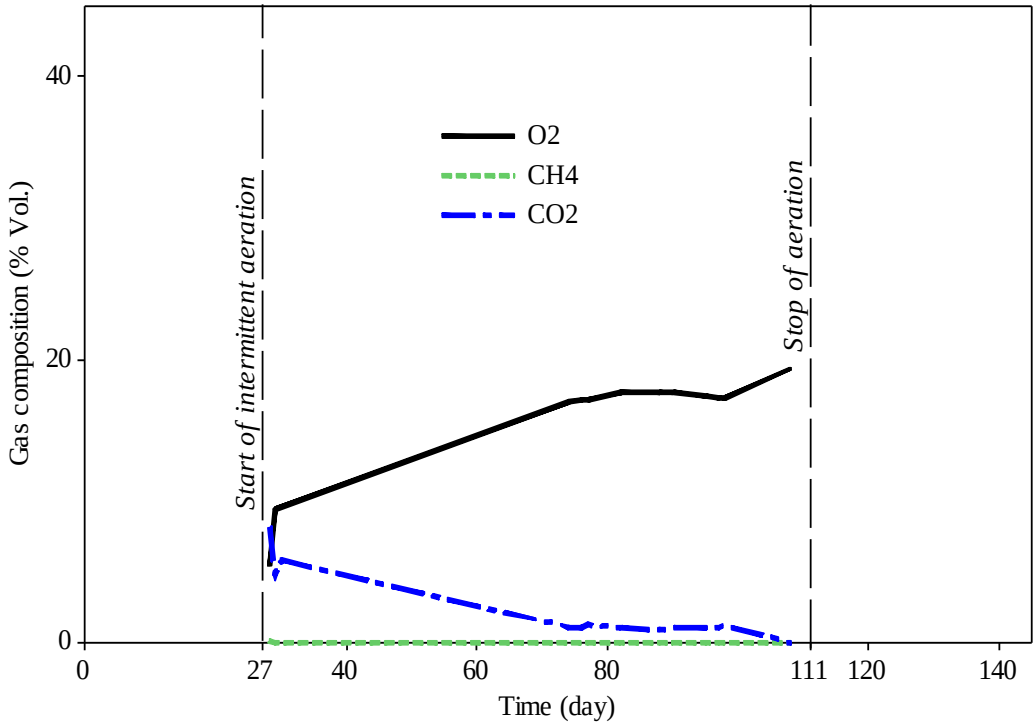


Figure 6.14. Variations in biogas composition from LSR 2

It can be seen from Figures 6.15 and 6.16 that the behaviour of the one-year old MSW (in lysimeters 3 and 4) differs from the MBT residue (in lysimeters 1 and 2) as well as the fresh MSW (in lysimeters 5 and 6). The change in biogas composition from MSW

lysimeter 3 after shifting the intermittent aeration to continuous regime is not expected due to a reduction of O_2 to 0 % and build-up of CH_4 is observed. This uncommon phenomenon may be attributed to the change in preferential flows caused by the continuous aeration.

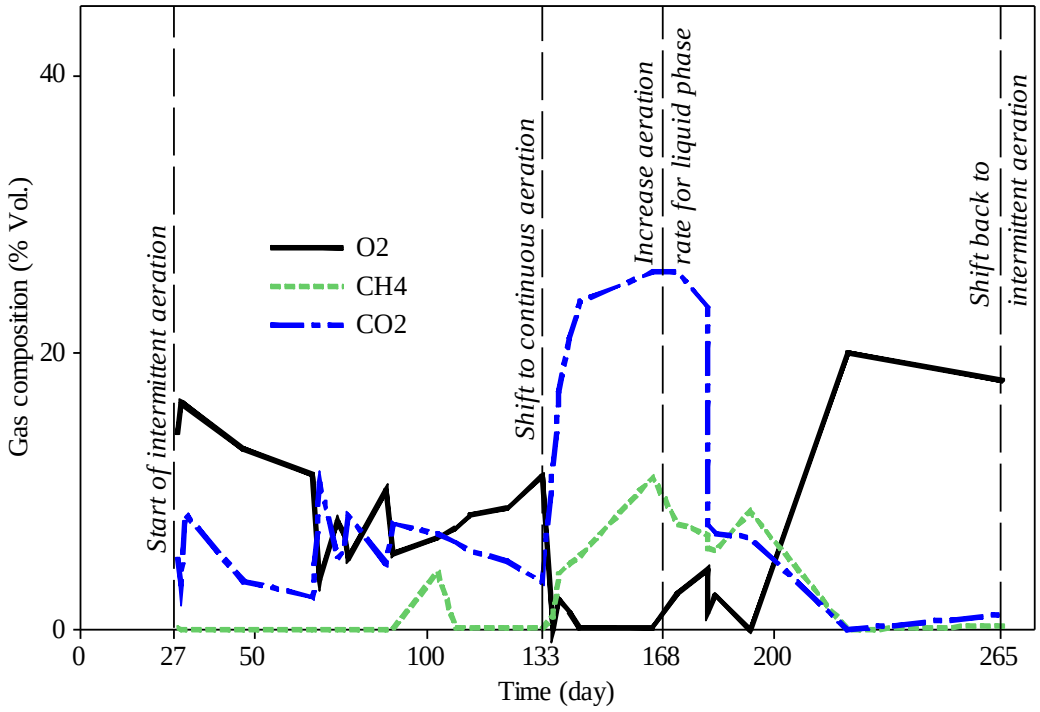


Figure 6.15. Variations in biogas composition from LSR 3

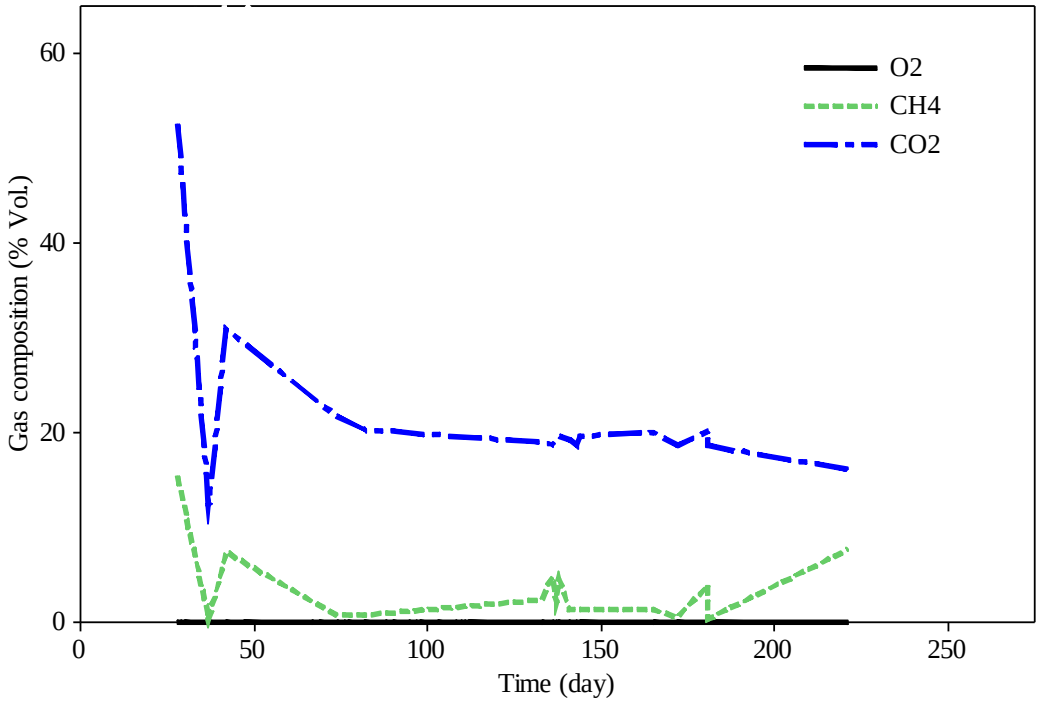


Figure 6.16. Variations in biogas composition from LSR 4

CO_2 and CH_4 levels in the emitted gas from the non-aerated MSW lysimeter 4 highly fluctuate in the first 2 months, then CO_2 values stabilise at about 20 %, whereas CH_4 values are very low. The pH value of the leachate from lysimeter 4 most of the time stabilised at around 6.0. Such condition could inhibite the activities of methanogenic bacteria. There was a very slight increase in pH values after 6 months of the operation. It can be seen that Figure 6.15 shows a small onset of methanogenic phase regarding lysimeter 4.

As for two aerated lysimeters 5 and 6 containing fresh MSW, the emitted gas from both lysimeters mainly included CO_2 (around 67 % for lysimeter 5 and 80 % for lysimeter 6) and small amount of CH_4 (around 5 % for lysimeter 5 and 12 % for lysimeter 6) prior they were aerated (see Figures 6.17 and 6.18).

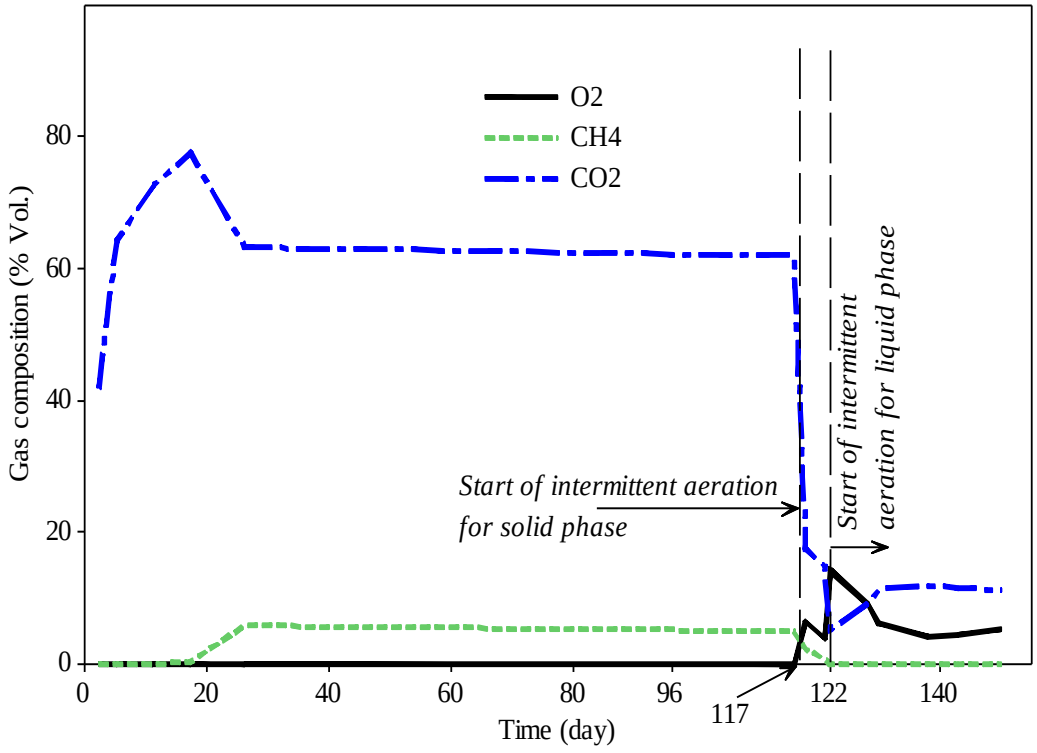


Figure 6.17. Variations in biogas composition from LSR 5

The pH value of about 6.0 in the leachates from both lysimeters 5 and 6 before the aeration was the reason for the inhibition of methanogenic bacteria in these lysimeters. It is also clear to see that under anaerobic conditions, the fresh MSW in lysimeters 5 and 6 under warm condition of about 30 °C has experienced a stronger degradation in comparison to the one-year old MSW in lysimeter 4 (comparing Figures 6.17 and 6.18 with Figure 6.16). As soon as the aeration was taking place in two lysimeters 5 and 6, CO_2 and CH_4 started to decrease.

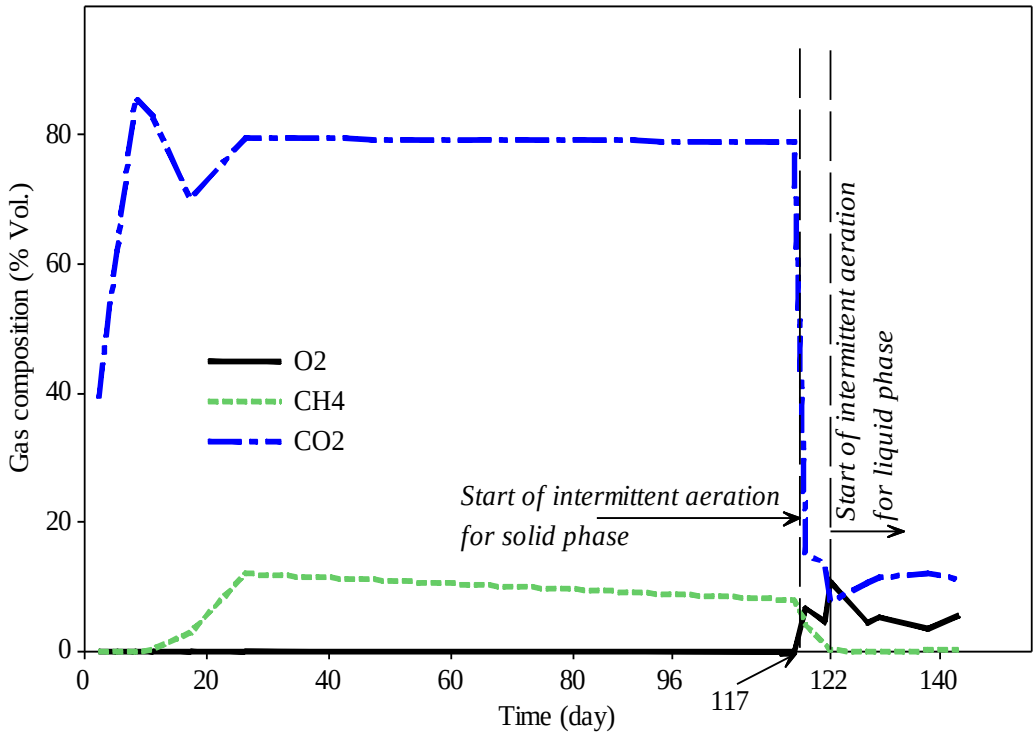


Figure 6.18. Variations in biogas composition from LSR 6

6.8 Evaluation of nitrogen and organic carbon removal

Nitrogen and carbon removal efficiencies are calculated from the experimental results regarding the leachates from the test lysimeters (Table 6.2).

For each lysimeter, nitrogen removal is evaluated by comparing the highest and ending values of ammonium, nitrite, nitrate, and total nitrogen in the leachate (the ending values were measured at the point of time as the operation was quit). In a similar manner, C removal is assessed by differing the peak and final values of TOC, COD, and VFA in the leachate. The evaluation does not include the MSW lysimeter 4 because it has experienced nearly no changes in the nitrogen and carbon parameter during the whole test period.

It can be clearly seen from Table 6.2 that the removal efficiency for total nitrogen in the leachate from the MBT lysimeter 1 is very elevated (above 98 %); meanwhile the C removal efficiency is relatively high at about 70 %. In the leachate from the MBT lysimeter 2, ammonium and nitrite nitrogen removal efficiencies were similarly as high as those values in case of lysimeter 1; however, the removal efficiency for nitrate nitrogen is not as high with 61.90 %, leading to the lower total nitrogen removal efficiency of only 72.93 % (compared with 98.48 % regarding the MBT lysimeter 1). TOC removal efficiency was in the same range as that of lysimeter 1, while the removal efficiencies for

COD and VFA were at 77.00 % and 82.76 %, fairly higher than those for lysimeter 1 which were at 69.53 % and 70.41 %, respectively.

Table 6.2. Nitrogen and carbon removal efficiencies regarding the leachates from aerated lysimeters in phase 3

Lysimeter		NH_4-N	NO_3-N	NO_2-N	TN	TOC	COD	VFA
MBT LSR 1	Max value	578	207.7	150.4	729.5	406	1513	176.4
	End value	2	3.4	0.3	11.1	116	461	52.2
	% removal	***99.65***	***98.36***	***99.80***	***98.48***	***71.43***	***69.53***	***70.41***
MBT LSR 2	Max value	582	413.1	553.2	703.4	361	1543	298.1
	End value	3	157.4	0.3	190.4	108	355	51.4
	% removal	***99.48***	***61.90***	***99.95***	***72.93***	***70.08***	***77.00***	***82.76***
MSW LSR 3	Max value	3518	*Not calculated due to very stable and low NO_3-N values*	*Not calculated due to NO_2-N under the detection limit*	5155	58459	217265	76411
	End value	1244			1847.6	3326	9569	3860
	% removal	***64.64***			***64.16***	***94.31***	***95.60***	***94.95***
MSW LSR 5	Max value	5300			6342	52094	180292	70667
	End value	1980			2745	7534	22009	10594
	% removal	***62.64***			***56.72***	***85.54***	***87.79***	***85.01***
MSW LSR 6	Max value	5527			6115	50826	187087	73566
	End value	1694			2732	4938	15945	4593
	% removal	***69.35***			***55.32***	***90.28***	***91.48***	***93.76***

The removal efficiencies for ammonium nitrogen, TOC and COD in the leachates from two MBT lysimeters in this phase and from two small 1.34-L MBT lysimeters in phase 2 (Chapter 5) are in similar range, regardless of the difference in dimension scale of the lysimeters as well as in aeration rate used. The volume of two large MBT lysimeters in this phase (141-L) is around 105 times larger than that of two small 1.34-L MBT lysimeters in phase 2, however, the diameter of aeration tube is also much bigger than the one installed in two small 1.34-L MBT lysimeters. Two large MBT lysimeters in this phase also have lower aeration rate compared with two small 1.34-L MBT lysimeters in

phase 2. Therefore, such mentioned removal efficiencies seem to be independent of the dimension scale of lysimeters as well as the aeration rate.

The experimental results for the concerned paramerers in the leachate from the MSW lysimeter 3 stated that the purposeful alterations in aeration regime (e.g. turning from intermittent to continuous, increase of the aeration rate for liquid phase) seemed to bring no significant changes to the values of such parameters. Therefore, intermittent aeration should be used whenever in situ aeration is intended to be applied.

The removal efficiencies for nitrogen and carbon parameters in the leachates from three MSW lysimeters LSR 3, 5, and 6 were in contrary to those from the two MBT lysimeters 1 and 2. In which, the removal efficiencies for ammonium and total nitrogen were much lower than those for TOC, COD, and VFA.

Ammonium nitrogen, TN, TOC, COD, VFA, and SO_4^{2-} concentrations in the leachates from two fresh MSW lysimeters 5 and 6 appear to have decreasing trends during the aeration process. It can be assumed the concentration of these parameters would have even decreased more as the operation of these lysimeters was continued for a longer time. Therefore, it should be expected that the nitrogen and carbon removal efficiencies for such lysimeters would be higher in case of maintaining the aeration longer.

6.9 Summary

The experimental results clearly show that the combination of leachate recirculation and aeration give positive impacts on the leachate quality, especially in view of nitrogen and carbon removal, regardless of either MBT or MSW material being investigated.

All aerated lysimeters have experienced moderately to dramatically reductions in their leachate nitrogen and carbon concentrations. However, the impacting mechanism for nitrogen reductions in case of two MBT lysimeters 1 and 2 differs from that regarding three aerated MSW lysimeters 3, 5, and 6.

As for two MBT lysimeters 1 and 2, significant reduction of ammonium nitrogen concentrations was followed by dramatical increase in nitrite and nitrate nitrogen concentrations in their leachates. Such behaviours indicate that nitrification was taken place to these systems, regardless of either intermittent or continuous aeration was implemented. It can be pointed out that the stop of continuous aeration for MBT lysimeter 1 was at the right time (on day 133, after 37 days of applying aeration) since the observed ammonium, nitrite, and nitrate nitrogen values showed the completion of nitrification at this point. The rapid reduction of nitrate nitrogen concentration in its

leachate to a very low level of 3.54 mg/kg DM after quitting aeration and afterwards stabilising at around 2.77 mg/kg DM was a solid evidence that the established anaerobic condition did ease the denitrification process and make it complete. As for MBT lysimeter 2, ammonium and nitrite nitrogen concentrations in its leachate decreased to very low values at 3.5 and 0.2 mg/kg DM, respectively on day 82 (after 55 days of applying aeration). However, the intermittent aeration was not stopped, but rather intentionally kept continuing for next 29 days. Concentration of nitrate nitrogen in its leachate was also decreased, however, to a value of about 160 mg/kg DM on day 221 and then kept stabilising at this value until the end of its operation. Due to this downtrend of nitrate nitrogen, total nitrogen values experienced a similar decreasing tendency as well. Such behaviour addresses that denitrification also occurred to the system, but was not completed. And in this case, nitrate nitrogen accounts for approximately 80 % of total nitrogen in the leachate.

Regarding the three aerated MSW lysimeters 3, 5, and 6, there were also significant reductions of ammonium and total nitrogen concentrations in their leachates, however, at lower degrees in comparison to those in the leachates from two MBT lysimeters. These MSW lysimeters experienced nearly no changes regarding nitrate nitrogen concentration in their leachates, while nitrite nitrogen was under the detection limit. Such mentioned behaviours and the increases in pH values of their leachates most likely suggest that nitrogen was lost via volatilisation, instead of either nitrification or denitrification processes.

In considering TOC, COD, and VFA concentrations in the leachates, the aeration applied to three MSW lysimeters appeared to have larger influences on the removal of such parameters compared with two MBT lysimeters. This should be due to the fact that the leachates from MSW lysimeters have significant proportion of easily biodegradable organic matters, whereas the leachates from MBT ones almost comprise hardly degradable organic compounds. The easy biodegradable organic compounds should have been successfully removed during MBT process. It is clearly observed that the highest TOC, COD, and VFA values in the leachates from MBT lysimeters are even though much lower than such lowest values from MSW lysimeters at the end of their operation.

As for the MSW lysimeter 4 operated under anaerobic condition, nitrite nitrogen concentration in its leachate was under the detection limit, meanwhile all other testing parameters were nearly stable throughout the whole test period. It is most likely that the lysimeter has been in “sour” state that anaerobic degradation was totally inhibited.

CHAPTER 7: CONCLUSIONS, RECOMMENDATIONS AND OUTLOOKS

7.1 Conclusions

By setting different boundary conditions for the operation of lab-scale lysimeters, the behaviours of leachates produced from such lysimeters have been investigated and compared.

In phase 1, the application of leachate recirculation solely has had little influence on improving the quality of leachates released from both types of the waste material during the experimental period.

With respect to the two MBT lysimeters, all testing parameters were stable before the temporarily malfunction of the lysimeters. Their leachate pH values were very slightly affected by this short breakdown. However, significant decreases in their leachate EC values, ammonium nitrogen, TOC, and COD concentrations were observed following this event. As the water addition for the compensation of leachate completed, these parameters achieved stable values again. Such mentioned reductions should have been primarily due to the strong flushing effect, caused by the unexpected dilutions.

Regarding the two fresh MSW lysimeters, their leachate ammonium nitrogen, TOC, and COD concentrations increased fast from the commencement to the end of their operation. However, the increase in these parameters' concentrations was elevated at the start-up and became gradual throughout their operational progress. The addition of water (after each sampling) seemed to make no reduction effect on the concentrations of such parameters, as well as the EC values, although it led to dilution effect at certain extent. This should be attributed to the fact that the flushing and/or leaching process was continuously taken place.

In phase 2, the combining technique of leachate recirculation and intermittent aeration has brought to a marked improvement in the quality of leachate from small lab-scale lysimeters containing residues from an MBT plant, especially in view of nitrogen removal. It was also the first time the combination technique has been applied to MBT residue. The utilisation of such technique has resulted in a fast and significant decrease in ammonium nitrogen and TKN, followed by a considerably rapid increase, and then decrease in nitrite and nitrate nitrogen concentrations. These changing behaviours imply that either nitrification was taken place in the system, followed by denitrification or both processes were alternatively functioned. The reduction in TN also addresses that certain N-containing gases were emitted from the system. At the end of the tests, the removal

efficiency for TN in the leachates from two hybrid lysimeters was totally high at around 99 %, whereas it was just about 6 to 15 % for the recirculation lysimeters without aeration. The experimental results show that the higher the aeration rate and time, the faster nitrogen conversion.

It was also observed a moderate reduction in TOC and a relatively fast decrease in COD concentrations in the leachates from the hybrid lysimeters. However, the reduction effect is not as strong as for nitrogen compounds and the intermittent aeration seems to be effective only at the first stage of aeration. The aeration intensity has only little influence on the decrease of TOC and COD. In case of the recirculation lysimeters, the slight decrease in TOC and COD was mainly caused by flushing and leaching effects.

Based on the experimental results of phase 2, it can be concluded that the combination of intermittent aeration and leachate recirculation has brought to significant reductions in concentrations of ammonium nitrogen (therefore also TKN) and relative small reductions of organic matters in the leachate from the hybrid lysimeters. Small reductions in such nitrogen parameters as for the recirculation lysimeters were also primarily due to flushing and leaching effects.

As for phase 3, the experimental results clearly show that the combination of leachate recirculation and aeration give positive impacts on the leachate quality, especially in view of nitrogen and carbon removal, not only in case of MBT residue, but also for either 1-year old MSW or fresh MSW material being investigated. The results also state that as soon as two MSW lysimeters containing fresh MSW were operated in the warm chamber, there were dramatic increases in ammonium nitrogen (therefore TN), TOC, COD, and VFA concentration in their leachates. Such increases imply that warm temperature accelerates the decomposition of the waste.

All five aerated lysimeters have experienced moderately to dramatically reductions in their leachate nitrogen and carbon concentrations, depending on the type of material used and/or the boundary setting conditions of the lysimeters. However, the nitrogen reductions in case of two MBT lysimeters bear a totally different nature and mechanism in comparison to those regarding three aerated MSW lysimeters.

As for two MBT lysimeters, significant reduction of ammonium nitrogen concentrations was followed by dramatical increase in nitrite and nitrate nitrogen concentrations in their leachates. These changes were very similar to what happened in the two small scale hybrid lysimeters in phase 2. Such behaviours one more time indicate that nitrification was taken place to these systems. These changes also imply that the nitrification took place, regardless of either intermittent or continuous aeration was implemented. In this

phase, it is essential to monitor the progress of nitrification to recognise the right point of time when it completes in order to stop the aeration, and also to facilitate the sequent denitrification process. By doing so, a high efficient removal of ammonium, nitrite, nitrate nitrogen, and therefore total nitrogen from the leachates can be achieved.

In three aerated MSW lysimeters, there were also significant reductions of ammonium and total nitrogen concentrations in their leachates, however, at lower degrees in comparison to those in the leachates from two MBT lysimeters. Additionally, these MSW lysimeters experienced nearly no changes in nitrate nitrogen concentration in their leachates, while nitrite nitrogen was under the detection limit. A combination of such mentioned behaviours and the increases in pH values of their leachates addresses that nitrogen reductions most likely attributed to the volatilisation of ammonia gas, other than either nitrification or denitrification processes.

In considering organic carbon parameters, including TOC, COD, and VFA in the leachates, the introduction of aeration to three MSW lysimeters appeared to have larger influences on the removal of such parameters in comparison to the two MBT lysimeters. This is due to the fact that the leachates from MSW lysimeters own a significant proportion of easily biodegradable organic matters, whereas the leachates from MBT ones almost comprise hardly degradable organic compounds. The easy biodegradable organic compounds should have been successfully removed during MBT process. It is also obvious that the highest TOC, COD, and VFA values in the leachates from MBT lysimeters are even though much lower than such lowest values from MSW lysimeters at the end of their operation.

As for the MSW lysimeter operating under anaerobic condition, nitrite nitrogen concentration in its leachate was under the detection limit, meanwhile all other testing parameters were nearly stable throughout the whole test period. It is most likely that the lysimeter has been in the “sour” state, leading to the inhibition of anaerobic degradation in the system.

The achieved results suggest that an additional stage of operation can considerably influence the long-term behaviour of the landfill. Such operation can be applied to MBT residues before or after their emplacement on landfills, as well as to MSW landfills. The additional treatment by either intermittent or continuous aeration in combination with leachate recirculation reduces the nitrogen concentrations in leachate to low and acceptable levels. The effort, costs and duration of leachate treatment in the aftercare phase can therefore be reduced considerably.

The achieved results also contribute to encourage developing countries utilising the abovementioned combining technique in accelerating the stabilisation of MSW landfills.

7.2 Recommendations and outlooks

Despite the fact that the aerated MSW lysimeters have achieved significant reductions in the concentrations of the concerned parameters (NH_4-N, TN, TOC, COD, VFA) in their leachates, the end values of such parameters (at the point of time their operation was terminated) were yet much higher than those in the leachates from MBT lysimeters. By aeration, the reduction of ammonium nitrogen in the leachate from MSW lysimeters was most likely due to the volatilisation of free ammonia, other than by nitrification. It means that nitrification was inhibited as applying aeration to MSW lysimeters. Therefore, further studies on inhibition factors should be done in the future.

Although the tests were conducted over more than 250 - 295 days (phase 1), 85 days (phase 2), and 310 - 413 days (phase 3), time is not sufficient to evaluate the long-term behaviour of the concerned parameters. This is particularly the case for the MSW lysimeters, in which ammonium nitrogen, TN, TOC, COD, and VFA concentrations were showing decreasing trends during the combination of aeration and leachate recirculation. Longer aeration should be carried out to the aerated MSW lysimeters to see whether:

- Ammonium nitrogen, TOC, COD, VFA concentrations in their leachates decrease to very low levels, like in case of MBT lysimeters;
- Any significant changes in nitrite and nitrate nitrogen occur;

Also regarding the aerated MSW lysimeters, it is suggested that the reduction of ammonium nitrogen in their leachates is caused by the volatilisation of ammonia gas. Therefore, their exhaust gas stream need to be regularly passed through an acid scrubber, especially when observing a decrease in ammonium nitrogen and an increase in pH values, to identify the variations in ammonia gas concentration.

After stopping aeration, the tests should be continued for all lysimeters in a longer stage, as it has been recommended by Fellner and Laner (2011), to observe their emission behaviours. It can be expected that the conditions inside such lysimeters will turn back to anaerobic with time. Concentrations of organic carbon and nitrogen compounds in their leachates may then increase again, which need to be observed. The time necessary for observation of this stage should be dependent on the boundary conditions of the tests as well as the expected values at the end of the operation.

Further studies should also have a closer look at the aerating phase, especially to MBT lysimeters as significant changes in ammonium, nitrite, and nitrate nitrogen of their leachate concentrations happen. Their gaseous phase should be analysed to specify emitted nitrogen-containing gases in more details.

In lab-scale tests, the recirculated leachate and the air ingress by aeration are quite easy to be evenly dispensed all over the waste due to the small dimension of the lysimeters and the relative homogeneity of the waste used. However, in field scale of both MBT and MSW landfills, the volume of deposited refuse is many times higher and much more inhomogenous. As a consequence the number of tube to be installed for aeration/irrigation is much higher and even then an uniform distribution of air/leachate is not guaranteed. The reason for this constraint is the formation of preferential flow paths, which means that in some parts of the waste volume air or water can flow very well, while in other parts no flow at all occurs. The formation of this dual porosity has a large influence on the efficiency of the in-situ treatment and should be tested before installation.

The experimental results suggest that it is possible to use the combination of aeration and leachate recirculation as an additional treatment step for MBT residue before it is deposited on landfills. The combination technique should, however, be adapted to the specific configurations of the MBT process. Alternatively it can be integrated into aerobic stabilisation by making this stage either more intensive or longer. This should be the case for developed countries where the direct landfilling of MSW has been banned. The technique can also apply to MBT landfills. However, aeration of MBT residue after emplacement on landfills should result in low outcome because the highly compacted MBT residue can greatly obstruct the distribution of air injected.

In developing countries where most of MSW is commonly disposed of in landfills without any treatment, the combination technique can be applied to accelerate the conversion and stabilisation of solid waste and enhance the leachate quality.

The very low levels of nitrogen and organic carbon parameters in the leachate from MBT residue compared with the high values of such parameters in the leachate from MSW prior to applying the combination technique have clearly addressed the advantages of MBT technologies over the conventional landfilling of MSW. Although the experimental results show positive outcomes as applying the combination technique on improving the quality of leachates generated from both MBT residue and fresh MSW, the direct landfilling of MSW without treatment (i.e. MBT) in any case should be avoided.

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APPENDICES

Appendix 1: The real settings of four 141-L lab-scale lysimeters for phase 1

Appendix 2: The real settings of four 1.34-L small lab-scale lysimeters for phase 2

Appendix 3: The real settings of 141-L lab-scale lysimeters 5 and 6 in the warm chamber for phase 3